JRC Scientific and Technical Reports

Polychlorinated biphenyls (PCBs) at the JRC Ispra Site: Air Concentrations, Congener Patterns and Seasonal variation

Results from the 1st year of atmospheric monitoring of persistent organic pollutants (POPs) at the Ispra EMEP station

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EUR 23258 EN - 2008

JRC EUROPEAN COMMISSION

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JRC 43258

EUR 23258 EN
ISSN 1018-5593

Luxembourg: Office for Official Publications of the European Communities

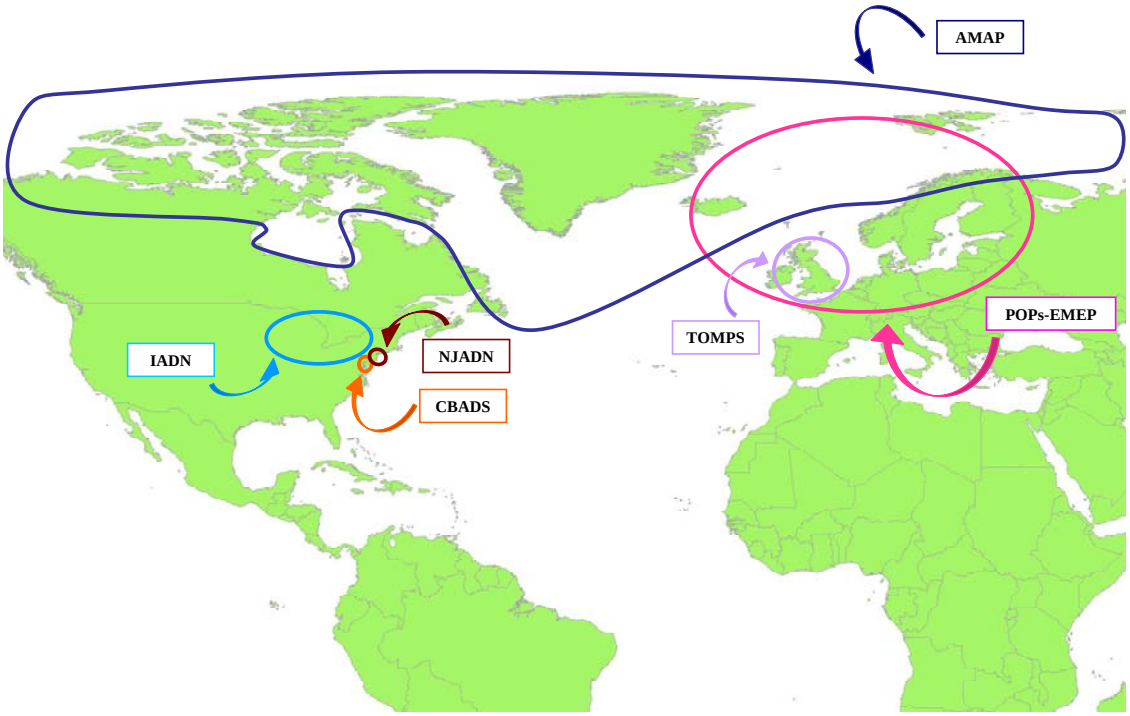
Printed in Italy

CONTENTS

1. Introduction and Objectives

The industrial revolution and the resultant technological society have led to a continuous production and emission of new toxic substances causing gradual and widely diffuse adverse effects to the entire planet usually known as Global Environmental Changes. Climate change, ozone depletion and global distribution of persistent organic pollutants (POPs) are, among other, important examples of detriment of global environmental quality. Different chemical families are considered as POPs, such as polychlorinated biphenyls (PCBs), a wide spectrum of organochlorine pesticides (OCPs), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polycyclic aromatic hydrocarbons (PAHs), and, polychlorinated naphthalenes (PCNs). In addition, some emerging contaminants are currently considered as candidate POPs, like the polybrominated diphenyl ethers (PBDEs) and the perfluorinated compounds (PFCs).

Several POPs monitoring programs are operative around the world (Figure 1). In the Artic region (eight circumpolar countries) the Artic Monitoring and Assessment Programme (AMAP) is measuring atmospheric concentrations of POPs since it was established in 1991. At a European scale, a major effort is being carried out under the UNECE Convection on Long-range Transboundary Air Pollution (CLRTAP), the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). This monitoring network was recently extended by a protocol on POPs. In addition, other POPs monitoring programmes exist at regional or national scales (eg. TOMPS in UK, NJADN in New Jersey-US, CBADS in Chesapeake Bay-US) and a large number of "independent" sites measuring atmospheric concentrations of POPs are spread out around the world. However, there are many "monitoring holes" and for numerous areas no information on POPs ambient levels is available. Such is the case with the EMEP network, that officially started to monitor POPs in 1999 but only a few sites are currently active within the network, leaving the Southern-Western Europe uncovered (Figure 2).



EMEP (Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe), **AMAP** (Arctic Monitoring and Assessment Programme), **TOMPS** (Toxic Organic Micropollutants) network , **IADN** (Integrated Atmospheric Deposition Network)**, NJADN** (New Jersey Atmospheric Deposition Network), **CBADS** (Chesapeake Bay Atmospheric Deposition Study)

Figure 1. Main POPs monitoring networks in Europe and North America.

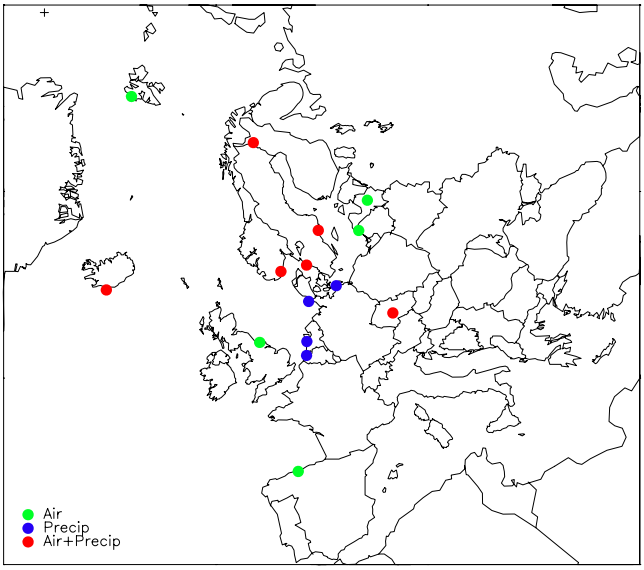


Figure 2. EMEP POPs monitoring sites, 2004 (from EMEP Status Report, 2006).

With the aim of gathering information on atmospheric POPs concentrations in one of the "monitoring holes" in Europe, a monitoring and research atmospheric site was set up at the EMEP Ispra station (see site description in section 2.1.). Regular monitoring activities started on April 2005 and finished on March 2007, in this first stage. Air and precipitation samples were collected regularly throughout this period. However, within the framework of other JRC research projects, previous POPs sampling was performed before entering into routine and results on PCDD/Fs, 7 ICES PCBs, dioxin-like PCBs (DL-PCBs) and PBDEs concentrations in air and precipitation and deposition fluxes from samples collected in the station in March 2005 have been already reported (Castro-Jiménez et al., 2008; Vives et al., 2007; Mariani et al., 2008).
The complete set of target compounds and a detailed overview of all sampling activities performed in the station can be found in Annexes I- III. Target POPs were selected on the bases on their existence in the main international POPs regulations such as the Stockholm Convention on POPs (UNEP, 2001) or the protocol on POPs to the CLRTAP (UNECE, 1998). This list was further extended by other relevant POPs for our research activities such as some pollutants from the Water Framework Directive Priority Substances List (European Commission, 2001).

The present report focuses on the first results obtained on PCBs ambient air concentrations from the first year of monitoring (April 2005-2006). PCBs are a group of 209 congeners of anthropogenic origin that enter the environment as a result of primary and secondary sources. PCBs were mainly used by the power industry in electrical transformers, capacitors, hydraulic equipment, and as lubricants. These compounds were also added to many products used directly by the public or the small size industry like adhesives, waxes and inks. Since the mid 1970s PCBs have been removed from active use in most countries. PCBs are present in almost all environmental compartments. Some current sources of emissions to the environment are landfills, open burning of products containing PCBs, waste incinerations and accidental fires (Breivik et al., 2002). Volatilization of PCBs from sites were they have been disposed or stored such as formerly exposed soils has also been reported as an important source (Cousins and Jones, 1998; Simcik et al., 1997). PCBs do bioaccumulate and are considered potent toxicants capable of producing a wide spectrum of adverse health effects in biota and humans such as induction of enzymes, anti-estrogenic effects, disruption of the endocrine system, immunosuppression, carcinogenicity, reproductive and developmental toxicity, skin disease, and cognitive disorders (Safe, 1984, 1990).

PCBs exist in the atmosphere as gases and bound to particles. Total airborne PCBs are normally dominated by the gas-phase burden (Mandalakis et al., 2002; Totten et al., 2004) temperature being an important variable affecting this partition (Schwarzenbach et al. 2003). This affinity to gas or

particulate phase is of relevant importance in the processes of atmospheric global transport and degradation (Pankow and Bidleman, 1992; Cotham and Bidleman, 1995). Among all PCB congeners, two groups are more frequently studied: (1) The 7 ICES (International Council for the Exploration of the Sea) PCBs, also known as indicator PCBs, since they are ubiquitous in all environmental compartments (CB-28, 52, 101, 118, 153, 138 and 180). (2) The so called dioxin-like PCB (CB-81, 77, 126, 169, 105, 114, 118, 123, 156, 157, 167, and 189). These congeners exhibit a very similar toxicity mechanism to dioxins due to certain molecular characteristics, which make them similar to the 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD), such as the fact that they can adopt a co-planar configuration (Landers and Bunce, 1991).

In the present work we report ambient air concentrations and congener pattern for the 7 ICES PCBs. In addition, the air gas/particle phase partitioning and the seasonal variation of the studied congeners are presented.

2. Materials and methods

2.1. Air sampling

Air samples were collected at the Joint Research Center EMEP Station at Ispra, Italy. This monitoring and research station operates under the Cooperative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). The sampling site is located in a semi-rural area by the Eastern shore of the sub-alpine Lake Maggiore, Lombardy, Northern Italy (45°49'N, 8°38'E, 209 m a.s.l) (Figure 3). The station is several tens of km away from large emission sources like intense road traffic or big factories. Milan (1,300 000 population), 60 km to the south-east; Novara (100 000 population), 40 km south; Varese (82 000 population), 20 km east; and Gallarate - Busto Arsizio (130 000 population), about 20 km south-east, are the main urban nucleus around.

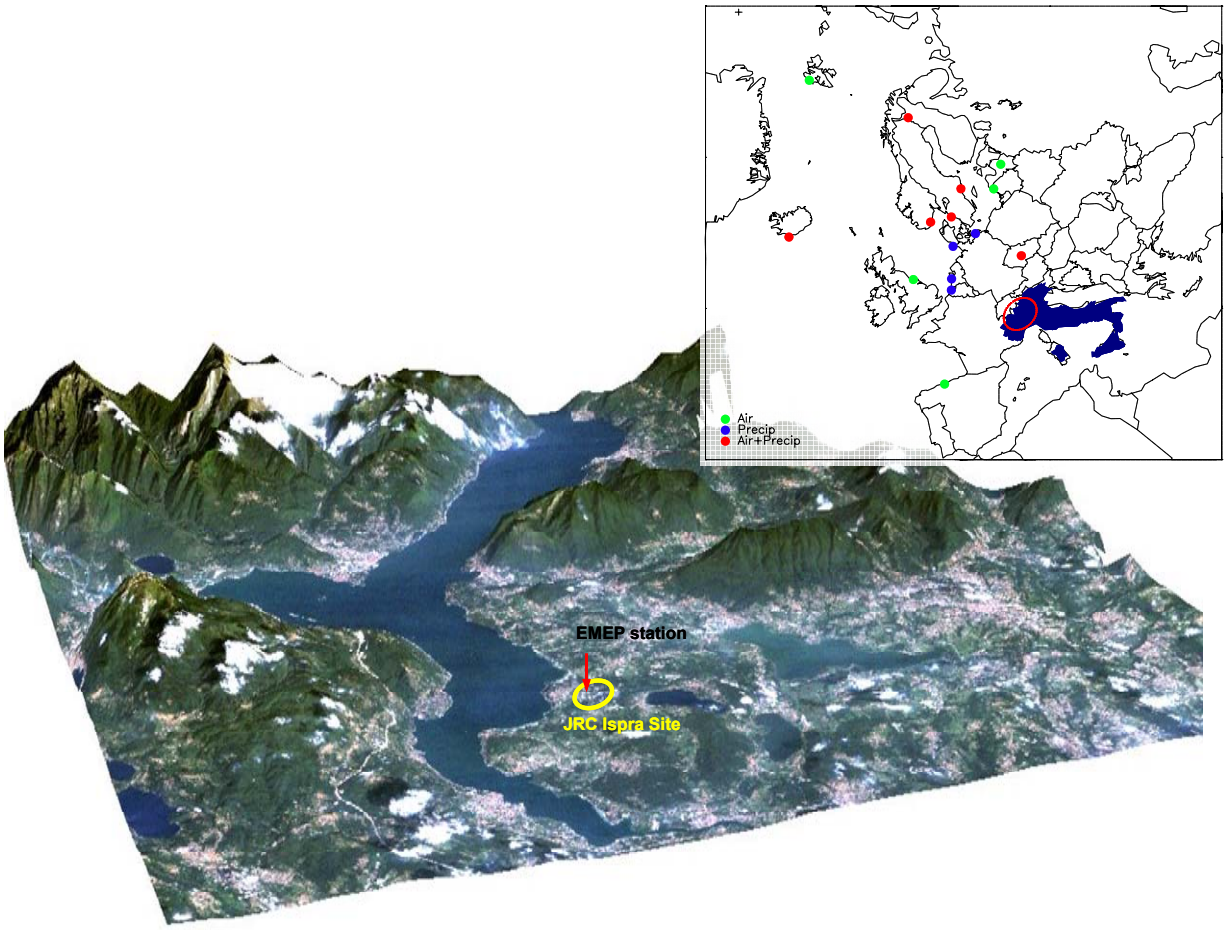


Figure 3. Location of the air sampling site at the EMEP station on the Eastern shore of Lake Maggiore (northern Italy).

Integrated weekly samples (one week resolution) were collected from April 2005 to April 2006 using high volume samplers (Echo PUF high volume sampler, TCR Tecora, Milan, Italy). Two air samplers were set up and were used alternatively to gather samples throughout the one year period. Air particle phase was retained by using a 102 mm diameter quartz fibre filter (QFF) from Whatman International Ltd (Brentford, Middlesex, UK) whereas the gas phase was trapped with a polyurethane foam (PUF) plug of 65 mm diameter, 75 mm length and 0.22 g/cm^{-3} in density. (Tisch Environmental, Inc., Cleves, Ohio, U.S.) (Figure 4). An average flow of 87 ± 2 L/min was achieved during the sampling interval gathering an averaged volume of 860 ± 27 m^3 for each sample. All details of sampling intervals are presented in Table 1.

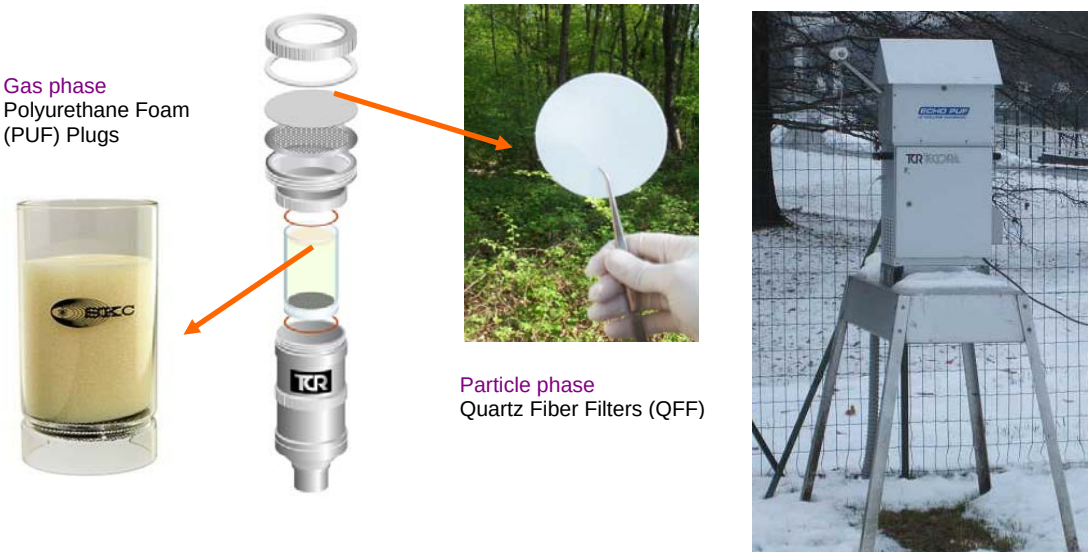


Figure 4. Detail of the sampling module, the material used, and the high volume sampler set up at the sampling station.

2.2. Analytical determinations

QFFs and PUFs were Soxhlet extracted separately with n-hexane/acetone (220:30 v/v) for 48 hours after being spiked with internal standards. PUFs were directly loaded into the soxhlet, whereas filters were first placed in extraction thimbles and then loaded into the soxhlet. Extract purification was executed with an automated clean-up system (Power-Prep P6, from Fluid Management Systems, Inc., Watertown, MA, USA). The purification method was previously described by Abad et al. (2000). Prior to injection, syringe ^{13}C-labelled standards were added to the extracts to determine the recoveries of the internal standards. Analysis of PCBs was based on isotope dilution using a high resolution gas chromatography – high resolution mass spectrometry (HRGC-HRMS). The GC (HP-6890, Hewlett Packard, Waldbronn, Germany), was coupled with a VG Autospec Ultima mass spectrometer (Micromass, Manchester, UK) operating in EI-mode at 34 eV with a resolution of >10000. Quantification was performed following 1668 U.S. EPA method (U.S. EPA., 1999). A HT-8 capillary column, 60 m long, 0.25 mm i.d. (inner diameter) and 0.25 µm film (SGE, Victoria, Australia) was used for the separation of the PCBs congeners.

MONTH	SAMPLE code	Start date	Finish date	Flow- l/min -	Vol (m^3)	Atm pressure (kPa)*	At. Av T (^oC)*
Apr-05	EMEP-2	30-Mar-05	6-Apr-05	86.980	869.216	99.870	12.4
Apr-05	EMEP-3	6-Apr-05	13-Apr-05	86.989	864.515	98.667	12.0
Apr-05	EMEP-4	14-Apr-05	21-Apr-05	86.984	869.270	97.254	10.1
Apr-05	EMEP-5	21-Apr-05	28-Apr-05	86.988	834.926	98.515	12.1
May-05	EMEP-6	29-Apr-05	6-May-05	86.986	856.338	98.692	17.5
May-05	EMEP-7	6-May-05	13-May-05	86.987	836.364	98.348	14.9
May-05	EMEP-8	13-May-05	20-May-05	86.987	848.821	98.579	15.0
May-05	EMEP-9	23-May-05	30-May-05	86.989	869.203	99.543	22.2
Jun-05	EMEP-10	30-May-05	6-Jun-05	86.988	868.880	99.179	22.8
Jun-05	EMEP-11	15-Jun-05	22-Jun-05	86.989	869.146	99.477	25.0
Jun-05	EMEP-12	22-Jun-05	30-Jun-05	86.989	869.249	99.211	26.6
Jul-05	EMEP-13	30-Jun-05	7-Jul-05	85.479	854.153	98.617	22.9
Jul-05	EMEP-14	8-Jul-05	15-Jul-05	86.988	869.179	99.053	22.1
Jul-05	EMEP-15	15-Jul-05	22-Jul-05	86.988	869.213	98.841	24.3
Jul-05	EMEP-16	22-Jul-05	29-Jul-05	86.990	869.125	98.870	25.0
Ago-05	EMEP-17	29-Jul-05	5-Aug-05	86.990	869.239	98.656	23.7
Ago-05	EMEP-18	5-Aug-05	11-Aug-05	102.800	876.517	98.798	20.9
Ago-05	EMEP-19	11-Aug-05	18-Aug-05	86.988	869.257	98.615	20.8
Ago-05	EMEP-21	24-Aug-05	31-Aug-05	86.988	869.241	99.070	20.9
Sep-05	EMEP-22	31-Aug-05	7-Sep-05	86.989	869.281	99.323	22.3
Sep-05	EMEP-23	7-Sep-05	14-Sep-05	86.990	869.288	98.936	17.0
Sep-05	EMEP-25	21-Sep-05	28-Sep-05	86.991	869.294	99.221	15.4
Oct-05	EMEP-26	28-Sep-05	5-Oct-05	86.990	869.289	99.054	12.0
Oct-05	EMEP-27	5-Oct-05	12-Oct-05	86.990	869.286	99.568	11.8
Oct-05	EMEP-28	12-Oct-05	19-Oct-05	86.990	869.262	99.590	8.9
Oct-05	EMEP-29	20-Oct-05	28-Oct-05	88.470	861.885	99.139	13.5
Nov-05	EMEP-30	28-Oct-05	4-Nov-05	86.990	869.291	99.492	9.6
Nov-05	EMEP-31	4-Nov-05	11-Nov-05	86.989	869.059	99.789	8.4
Nov-05	EMEP-32	11-Nov-05	18-Nov-05	86.988	869.262	98.526	8.6
Nov-05	EMEP-33	18-Nov-05	25-Nov-05	86.990	869.312	98.911	-2.5
Dec-05	EMEP-34	25-Nov-05	2-Dec-05	86.989	869.302	97.249	1.0
Dec-05	EMEP-35	2-Dec-05	9-Dec-05	88.654	872.815	97.934	-1.9
Dec-05	EMEP-36	9-Nov-05	16-Dec-05	86.988	869.276	99.154	1.5
Dec-05	EMEP-37	16-Dec-05	23-Dec-05	86.990	852.411	98.760	-5.1
Dec-05	EMEP-38	23-Dec-05	30-Dec-05	86.989	869.273	97.969	0.0
Jan-06	EMEP-39	3-Jan-06	10-Jan-06	86.991	869.192	99.183	-3.2
Jan-06	EMEP-40	10-Jan-06	17-Jan-06	86.986	869.153	99.598	-1.9
Jan-06	EMEP-41	17-Jan-06	24-Jan-06	86.933	703.083	98.745	-3.7
Jan-06	EMEP-42	24-Jan-06	31-Jan-06	86.988	869.263	99.085	0.5
Feb-06	EMEP-44	7-Feb-06	14-Feb-06	86.989	869.330	98.392	2.0
Feb-06	EMEP-45	14-Feb-06	21-Feb-06	86.989	869.197	97.699	3.0
Feb-06	EMEP-46	21-Feb-06	28-Feb-06	86.99	864.4	98.014	1.1
Mar-06	EMEP-47	28-Feb-06	7-Mar-06	86.989	869.252	97.344	4.8
Mar-06	EMEP-48	7-Mar-06	14-Mar-06	86.991	869.328	98.055	2.5
Mar-06	EMEP-49	14-Mar-06	21-Mar-06	86.988	869.267	98.39	6.8
Mar-06	EMEP-50	21-Mar-06	28-Mar-06	86.989	788.495	98.286	7.5
Apr-06	EMEP-51	28-Mar-06	4-Apr-06	86.989	869.181	98.636	9.3
Apr-06	EMEP-52	4-Apr-06	11-Apr-06	86.989	869.299	98.009	10.4
Apr-06	EMEP-53	11-Apr-06	18-Apr-06	86.989	869.223	98.366	13.0

: SAMPLING PERFORMED IN DUPLICATE (QA/QC)

* Averaged atmopsheric pressure and temperature during the sampling event. Values were recorded by the meteo sensor from the air sampler

Table 1. Air sampling details, average temperature and atmospheric pressure during the sampling period.

2.3. Quality assurance /Quality control

A strict quality assurance/ quality control (QA/QC) protocol was followed during the sampling and analysis. QFF were individually wrapped in aluminium foil, baked at 450 ºC for 6 h and then stored in the freezer in sealed plastic bags till they were used. PUFs were soxhlet extracted during at least 24 hours with acetone and then dried in a dissicator under vacuum. Once the plugs were dry, they were individually wrapped in n-hexane rinsed aluminium, and then stored in sealed plastic bags till they were used. Field blanks consisting on cleaned QFFs and PUF mounted in the sampling head,

transported to the sampling site, mounted in the sampler, dismounted and transported back to the laboratory, were collected regularly. A breakthrough test was also performed by placing a smaller PUF plug in series separated ~1 cm from the main PUF in the sampling head. In addition, since two high volume samplers were used alternatively, several samples were collected in parallel to check the reproducibility of individual sampling devices. Samples at different flow rates were also collected to check a potential influence of the flow rate in the sampling.

Soxhlet extraction blanks, consisting on only extraction solvent or soxhlet thimbles with extraction solvent, were run for each batch of samples. Standards (natives + ^{13}C-compounds) were introduced in the chromatographic sequence to evaluate possible variations during the time of analyses. Target compounds chromatographic peaks where only considered when complying with the following QA/QC criteria: (1) The retention time of target compound were ± 3s of those observed for the corresponding standards; (2) Experimental isotopic ratio of natives compounds were within ± 20% of the theoretical ratio. (3) Peaks were at least 3 times higher than the noise. The LOD was calculated on the bases of a signal to noise ratio of 3/1.

3. Results and discussion

3.1. QA/QC

Field Blanks

Several filed blanks were taken during the sampling period in order to check PCB background levels inherent to the sampling material or procedures. An average value (n=2) of 3 pg m^{-3} was obtained for the ∑ 7 PCBs (gas + particulate phase). This value represent ~ 10 % of the lowest concentration value obtained for the ∑ 7 PCBs, December 05 (the worst case scenario), and its only ~ 6 % of the average monthly value (Table 3). Therefore, concentrations reported were not corrected by field blank values.

Breakthrough

Results from the breakthrough test indicated that this artifact was not significant under the experimental conditions assayed. A ∑ 7 PCBs concentration of 1.3 pg m^{-3} was found in the breakthrough PUF placed after the main PUF (gas phase concentration).This values is very similar to the average (n=2) ∑ 7 PCBs gas phase concentration found in the field blanks (1.4 pg m^{-3}).

Parallel sampling

Air samples were gathered using two identical high volume samplers alternatively. In order to check the reproducibility of the sampling, 4 samples were taken in duplicate (in the months of May, June, July and October 2005). A good agreement was found between concentrations in the samples collected with two different samplers in the same period and place (Table 2).

∑ 7 PCB concentration (gas + particulate phase) in pg m^{-3} for the parallel sampling							
May-05		Jun-05		July-05		October-05	
EMEP-9A	EMEP-9B	EMEP-11A	EMEP-11B	EMEP-13A	EMEP-13B	EMEP-29A	EMEP-29B
66.6	**59.1**	**51.8**	**69.1**	**60.5**	**58.3**	**62.0**	**62.9**

Table 2. PCB concentrations obtained for the duplicated samples

Sampling flow rate influence

Occasionally, some adjustments had to be performed to compensate unforeseen situations while sampling. Sampler flow rate was increased in several samples to acquire desired volumes in shorter times in order to fit the sampling program. To assure comparability of samples a test, for checking a potential influence of different flow rates, was performed operating two samplers contemporaneously. One sampler was set up at the usually employed flow rate (87 lpm), whereas the other device was operated at 240 lpm (the maximum sampler flow rate) for a shorter time. A concentration of 68.0 pg m^{-3} for the ∑7 PCBs (gas + particulate phase) was obtained for the sampler at a higher flow rate, whereas a value of 67.2 pg m^{-3} was obtained for the lower flow rate sampler. Results indicated that there were no differences in the conditions assayed.

3.2. Ambient air concentrations

The monthly average concentrations for the ∑ 7 ICES PCBs (gas+ particulate phases) are reported in Figure 5, where a first indication of the yearly evolution can be also observed. Generally, four sampling events were collected and analyzed for each month except for the months of June, September, February 2005 and April 2006 where three event were collected and for the month of December 2005 where five events were collected (Table 1). Total and individual average monthly congener concentrations (gas + particulate phase) are presented in Table 3.

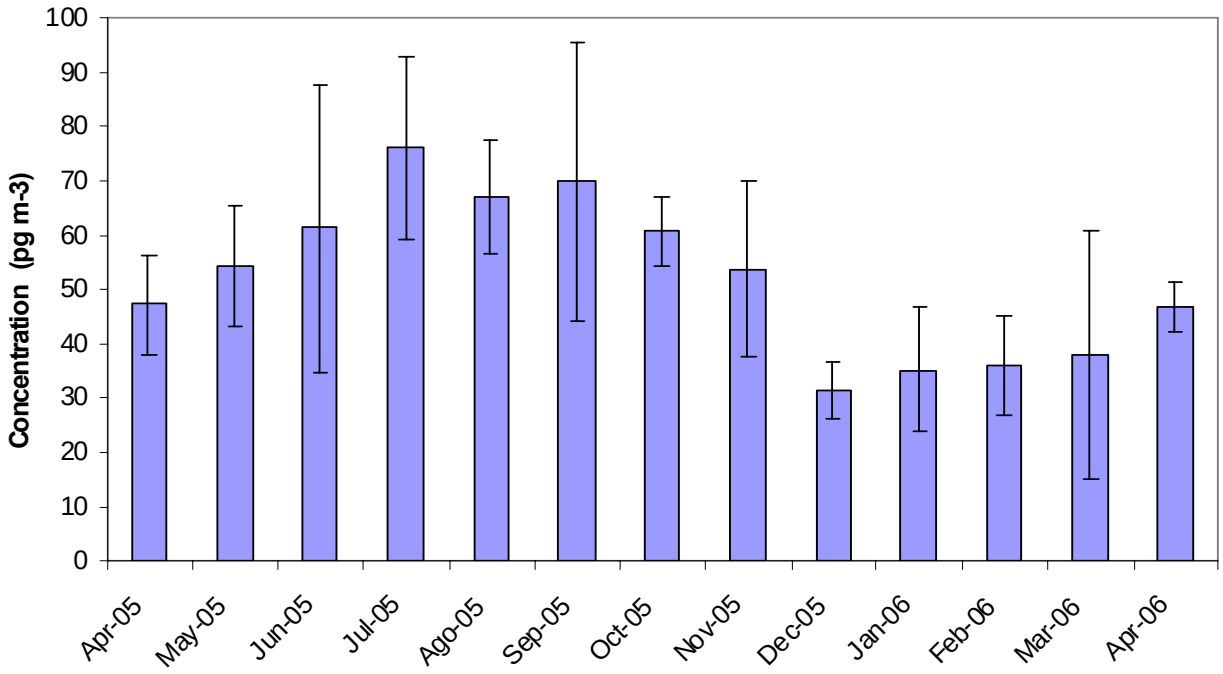


Figure 5. Monthly average concentrations (∑ 7 ICES, gas + particulate phases) for the sampling period (see Table 1 for details of the sampling event for each month). Bars are standard deviation.

The range of total (gas + particulate phase) monthly averaged concentration during the sampling period varied from 31 ± 5 to 76 ± 17 pg m^{-3}. The highest concentration corresponded to the month of July whereas the lowest to the month of December. Regarding individual congeners, a predominance of TeCB- 52 was observed, with a concentration ranging from 7 ± 2 to 22 pg m^{-3}, followed by CB-101 and CB-153, ranging from 5 ± 0.4 to 17 ± 4 and from 5 ± 0.7 to 16 ± 4 pg m^{-3}, respectively. Congener 28 ranged from 6 ± 0.5 to 12 ± 4 pg m^{-3} whereas PCBs 138, 118 and 180 varied from 3 ± 0.7 to 12 ± 4, from 3 ± 0.4 to 8 ± 1 and from 2 ± 0.8 to 7 ± 2 pg m^{-3}, respectively (Table 3).

Individual PCB congener and ∑ 7PCB concentrations in the air gas and particulate phases are presented in Table 4 and Table 5, respectively. Concentrations in the gas phase during the sampling period were higher than those in the particular phase. ∑ 7PCB gas phase concentrations ranged from 21 ± 2 to 72 ± 17 pg m^{-3} whereas ∑ 7PCB particulate phase concentration varied from 3 ± 0.5 to 10 ± 4 pg m^{-3}.

The concentrations were in the rage of those usually reported for rural, semi-rural or remote sites around the world (Axelman and Broman, 2001; Brunciak et al., 2001; Buehler et al., 2002; Mandalakis et al., 2001; 2002; 2007; Shen et al., 2006; Van Drooge et al., 2002; 2004; Yeo et al., 2004).

Montly average TOTAL concentration (gas + particulate phase) / pg m^{-3} ± SD (number of samples considered, n)								
Month	**TrCB28**	**TeCB-52**	**PeCB-101**	**PeCB-118**	**HxCB-153**	**HxCB-138**	**HpCB-180**	**∑ 7 PCBs**
Apr-05	10.5 ± 1.8 (*n=3*)	12.5 ± 1.5 (n=3)	9.7 ± 1.8 (n=4)	3.5 ± 0.7 (n=4)	7.6 ± 1.7 (n=3)	5.3 ± 1.2 (n=4)	3.8 ± 0.9 (n=4)	47.2 ± 9.2 (n=4)
May-05	9.5 ± 1.6 (*n=4*)	15.9 ± 3.7 (n=4)	10.3 ± 2.6 (n=4)	3.9 ± 1.1 (n=4)	7.1 ± 2.1 (n=4)	4.6 ± 1.3 (n=4)	3.2 ± 0.8 (n=4)	54.6 ± 11.1 (n=4)
Jun-05	9.2 ± (*n=1*)	22.5 ± (*n=1*)	17.5 ± 4.1 (n=3)	7.1 ± 2.5 (n=3)	13.0 ± 3.7 (n=3)	8.8 ± 3.0 (n=3)	4.4 ± 0.5 (n=3)	61.4 ± 26.5 (n=3)
Jul-05	10.1 ± 1.9 (*n=4*)	21.7 ± 5.5 (n=4)	16.1 ± 4.0 (n=4)	6.3 ± 1.8 (n=4)	11.4 ± 2.5 (n=4)	7.0 ± 1.5 (n=4)	3.7 ± 0.3 (n=4)	76.3 ± 16.8 (n=4)
Ago-05	10.2 ± 1.3 (*n=4*)	20.3 ± 3.7 (n=4)	13.8 ± 2.4 (n=4)	5.2 ± 1.0 (n=4)	9.1 ± 1.8 (n=4)	5.4 ± 1.0 (n=4)	2.8 ± 0.5 (n=4)	66.9 ± 10.4 (n=4)
Sep-05	11.0 ± (*n=1*)	15.9 ± (*n=1*)	13.6 ± 2.2 (n=2)	8.2 ± 1.5 (n=2)	16.1 ± 4.0 (n=2)	11.8 ± 4.4 (n=2)	6.8 ± 2.0 (n=2)	69.9 ± 25.7 (n=2)
Oct-05	11.5 ± 0.7 (*n=4*)	15.4 ± 1.4 (n=4)	10.7 ± 1.3 (n=4)	5.6 ± 0.9 (n=4)	8.6 ± 1.1 (n=4)	5.2 ± 0.9 (n=4)	3.7 ± 0.4 (n=4)	60.7 ± 6.4 (n=4)
Nov-05	12.0 ± 4.2 (*n=4*)	14.4 ± 5.4 (n=4)	9.0 ± 3.1 (n=4)	3.8 ± 1.0 (n=4)	6.6 ± 1.5 (n=4)	4.0 ± 0.7 (n=4)	3.9 ± 0.3 (n=4)	53.8 ± 16.1 (n=4)
Dec-05	5.7 ± 0.5 (n=3)	7.2 ± 0.8 (n=5)	5.5 ± 0.4 (n=5)	2.9 ± 0.4 (n=5)	5.1 ± 0.7 (n=5)	3.5 ± 0.7 (n=5)	3.8 ± 1.3 (n=5)	31.4 ± 5.1 (n=5)
Jan-06	5.7 ± 2.0 (n=3)	6.8 ± 2.1 (n=3)	6.8 ± 3.2 (n=3)	4.1 ± 2.2 (n=3)	5.5 ± 1.6 (n=3)	3.8 ± 0.9 (n=3)	2.7 ± 0.5 (n=3)	35.3 ± 11.2 (n=3)
Feb-06	8.2 ± 2.0 (n=3)	9.7 ± 1.2 (n=2)	5.9 ± 1.3 (n=3)	2.9 ± 0.4 (n=3)	5.2 ± 0.7 (n=3)	4.6 ± 1.4 (n=3)	2.8 ± 0.4 (n=3)	36.1 ± 9.0 (n=3)
Mar-06	9.6 ± 5.7 (n=3)	9.7 ± 5.7 (n=4)	6.8 ± 4.2 (n=4)	2.9 ± 1.6 (n=4)	5.4 ± 2.6 (n=4)	3.6 ± 1.4 (n=4)	2.4 ± 0.8 (n=4)	38.0 ± 22.6 (n=4)
Apr-06	9.2 ± 0.3 (n=3)	11.4 ± 1.1 (n=3)	8.8 ± 0.8 (n=3)	3.8 ± 0.6 (n=3)	6.8 ± 0.9 (n=3)	4.3 ± 0.7 (n=3)	2.5 ± 0.5 (n=3)	46.8 ± 4.7 (n=3)
Range	5.7 - 12.0	6.8 - 22.5	5.5 - 17.5	2.9 - 8.2	5.1 - 16.1	3.5 - 11.8	2.4 - 6.8	31.4 - 76.3

Table 3. Monthly average concentrations for individual PCB congeners and ∑ 7 PCBs. (Note that for some congener and in some cases the number of samples considered is lower than the samples collected. This is due to analytical problems for those specific congeners in the particular months considered.)

Montly average GAS phase concentration / pg m^{-3} ± SD (number of samples considered, n)								
Month	**TrCB28**	**TeCB-52**	**PeCB-101**	**PeCB-118**	**HxCB-153**	**HxCB-138**	**HpCB-180**	**∑ 7 PCBs**
Apr-05	10.4 ± 1.9 (n=3)	12.4 ± 1.6 (n=3)	9.3 ± 1.7 (n=4)	3.3 ± 0.6 (n=4)	6.6 ± 1.5 (n=4)	4.4 ± 1.0 (n=4)	2.3 ± 0.5 (n=4)	43.0 ± 9.1 (n=4)
May-05	9.3 ± 1.6 (n=4)	15.7 ± 3.7 (n=4)	9.9 ± 2.6 (n=4)	3.7 ± 1.1 (n=4)	6.1 ± 2.0 (n=4)	3.7 ± 1.2 (n=4)	1.9 ± 0.7 (n=4)	50.2 ± 10.7 (n=4)
Jun-05	9.0 ± (n=1)	22.3 ± (n=1)	17.0 ± 4.2 (n=3)	6.8 ± 2.5 (n=3)	12.0 ± 3.8 (n=3)	7.7 ± 3.1 (n=3)	2.9 ± 0.8 (n=3)	56.9 ± 27.2 (n=3)
Jul-05	9.8 ± 1.9 (n=4)	21.5 ± 5.5 (n=4)	15.7 ± 4.0 (n=4)	6.1 ± 1.8 (n=4)	10.5 ± 2.5 (n=4)	6.2 ± 1.5 (n=4)	2.6 ± 0.3 (n=4)	72.5 ± 16.9 (n=4)
Ago-05	10.0 ± 1.3 (n=4)	20.1 ± 3.7 (n=4)	13.4 ± 2.4 (n=4)	5.0 ± 1.0 (n=4)	8.3 ± 1.8 (n=4)	4.7 ± 0.9 (n=4)	1.8 ± 0.4 (n=4)	63.2 ± 10.8 (n=4)
Sep-05	10.8 ± (n=1)	15.8 ± (n=1)	13.2 ± 2.2 (n=2)	8.0 ± 1.4 (n=2)	15.3 ± 4.0 (n=2)	11.1 ± 4.5 (n=2)	5.9 ± 2.2 (n=2)	66.8 ± 25.9 (n=2)
Oct-05	11.2 ± 0.6 (n=4)	15.1 ± 1.4 (n=4)	10.2 ± 1.3 (n=4)	5.2 ± 1.0 (n=4)	7.4 ± 1.4 (n=4)	4.2 ± 1.1 (n=4)	2.2 ± 0.5 (n=4)	55.5 ± 7.0 (n=4)
Nov-05	11.7 ± 4.3 (n=4)	14.1 ± 5.5 (n=4)	8.4 ± 3.4 (n=4)	3.1 ± 1.3 (n=4)	4.9 ± 2.1 (n=4)	2.6 ± 1.1 (n=4)	1.5 ± 0.7 (n=4)	46.4 ± 18.2 (n=4)
Dec-05	5.5 ± 0.5 (n=3)	6.9 ± 0.8 (n=5)	4.6 ± 0.5 (n=5)	1.9 ± 0.3 (n=5)	2.7 ± 0.3 (n=5)	1.5 ± 0.2 (n=5)	0.6 ± 0.2 (n=5)	21.4 ± 2.3 (n=5)
Jan-06	5.2 ± 1.8 (n=3)	6.3 ± 1.9 (n=3)	5.6 ± 2.9 (n=3)	2.8 ± 1.8 (n=3)	3.1 ± 1.5 (n=3)	1.8 ± 0.8 (n=3)	0.6 ± 0.4 (n=3)	25.3 ± 10.3 (n=3)
Feb-06	7.5 ± 1.8 (n=3)	8.9 ± 1.2 (n=2)	5.0 ± 1.3 (n=3)	2.1 ± 0.4 (n=3)	3.2 ± 0.7 (n=3)	2.9 ± 1.3 (n=3)	0.8 ± 0.3 (n=3)	27.5 ± 8.2 (n=3)
Mar-06	9.2 ± 5.6 (n=3)	9.3 ± 5.7 (n=4)	6.1 ± 4.3 (n=4)	2.3 ± 1.5 (n=4)	3.8 ± 2.5 (n=4)	2.3 ± 1.4 (n=4)	1.1 ± 0.6 (n=4)	31.9 ± 22.1 (n=4)
Apr-06	8.8 ± 0.2 (n=3)	11.2 ± 1.1 (n=3)	8.4 ± 0.7 (n=3)	3.5 ± 0.5 (n=3)	5.9 ± 0.6 (n=3)	3.5 ± 0.5 (n=3)	1.7 ± 0.4 (n=3)	43.0 ± 3.8 (n=3)
Range	5.2 - 11.7	6.3 - 22.3	4.6 - 17.0	1.9 - 8.0	2.7 - 15.3	1.5 - 11.1	0.6 - 5.9	21.4 - 72.5

Table 4. Monthly average air gas phase concentrations for individual PCB congeners and ∑ 7 PCBs. (Note that for some congener and in some cases the number of samples considered is lower than the samples collected. This is due to analytical problems for those specific congeners in the particular months considered.)

Montly average PARTICULATE phase concentration / pg m^{-3} ± SD (number of samples considered, n)								
Month	**TrCB28**	**TeCB-52**	**PeCB-101**	**PeCB-118**	**HxCB-153**	**HxCB-138**	**HpCB-180**	**∑ 7 PCBs**
Apr-05	0.18 ± 0.05 (*n=3*)	0.18 ± 0.05 (*n=3*)	0.47 ± 0.20 (n=4)	0.27 ± 0.08 (n=4)	0.94 ± 0.24 (n=4)	0.91 ± 0.24 (n=4)	1.48 ± 0.47 (n=4)	4.3 ± 1.1 (n=4)
May-05	0.18 ± 0.05 (*n=4*)	0.22 ± 0.05 (n=4)	0.42 ± 0.05 (n=4)	0.24 ± 0.02 (n=4)	0.96 ± 0.13 (n=4)	0.94 ± 0.22 (n=4)	1.35 ± 0.32 (n=4)	4.3 ± 0.8 (n=4)
Jun-05	0.30 ± 0.09 (*n=3*)	0.28 ± 0.06 (*n=3*)	0.47 ± 0.07 (n=3)	0.28 ± 0.03 (n=3)	1.03 ± 0.17 (n=3)	1.05 ± 0.25 (n=3)	1.45 ± 0.41 (n=3)	4.9 ± 1.1 (n=3)
Jul-05	0.22 ± 0.05 (*n=4*)	0.23 ± 0.06 (n=4)	0.39 ± 0.05 (n=4)	0.23 ± 0.02 (n=4)	0.86 ± 0.06 (n=4)	0.78 ± 0.05 (n=4)	1.06 ± 0.06 (n=4)	3.8 ± 0.3 (n=4)
Ago-05	0.26 ± 0.10 (*n=4*)	0.24 ± 0.03 (n=4)	0.39 ± 0.11 (n=4)	0.23 ± 0.04 (n=4)	0.84 ± 0.29 (n=4)	0.75 ± 0.32 (n=4)	1.02 ± 0.50 (n=4)	3.7 ± 1.2 (n=4)
Sep-05	0.18 ± 0.03 (*n=3*)	0.17 ± 0.03 (*n=3*)	0.33 ± 0.01 (n=3)	0.25 ± 0.06 (n=3)	0.72 ± 0.08 (n=3)	0.63 ± 0.11 (n=3)	0.85 ± 0.27 (n=3)	3.1 ± 0.5 (n=3)
Oct-05	0.27 ± 0.03 (*n=4*)	0.26 ± 0.05 (n=4)	0.46 ± 0.09 (n=4)	0.43 ± 0.12 (n=4)	1.16 ± 0.37 (n=4)	1.02 ± 0.26 (n=4)	1.54 ± 0.23 (n=4)	5.1 ± 1.0 (n=4)
Nov-05	0.25 ± 0.08 (*n=4*)	0.29 ± 0.11 (n=4)	0.68 ± 0.27 (n=4)	0.72 ± 0.23 (n=4)	1.65 ± 0.53 (n=4)	1.41 ± 0.41 (n=4)	2.41 ± 0.56 (n=4)	7.4 ± 2.1 (n=4)
Dec-05	0.24 ± 0.04 (n=5)	0.33 ± 0.08 (n=5)	0.91 ± 0.30 (n=5)	1.03 ± 0.36 (n=5)	2.39 ± 0.88 (n=5)	2.00 ± 0.82 (n=5)	3.19 ± 1.45 (n=5)	10.1 ± 3.8 (n=5)
Jan-06	0.47 ± 0.21 (n=3)	0.57 ± 0.18 (n=3)	1.20 ± 0.32 (n=3)	1.36 ± 0.48 (n=3)	2.36 ± 0.30 (n=3)	1.98 ± 0.21 (n=3)	2.05 ± 0.13 (n=3)	10.0 ± 1.2 (n=3)
Feb-06	0.70 ± 0.22 (n=3)	0.71 ± 0.17 (n=3)	0.92 ± 0.09 (n=3)	0.84 ± 0.23 (n=3)	1.94 ± 0.43 (n=3)	1.69 ± 0.43 (n=3)	1.93 ± 0.62 (n=3)	8.7 ± 1.8 (n=3)
Mar-06	0.43 ± 0.19 (n=4)	0.40 ± 0.18 (n=4)	0.64 ± 0.17 (n=4)	0.56 ± 0.20 (n=4)	1.54 ± 0.45 (n=4)	1.33 ± 0.39 (n=4)	1.30 ± 0.47 (n=4)	6.2 ± 2.0 (n=4)
Apr-06	0.34 ± 0.06 (n=3)	0.25 ± 0.07 (n=3)	0.40 ± 0.10 (n=3)	0.32 ± 0.08 (n=3)	0.93 ± 0.37 (n=3)	0.84 ± 0.31 (n=3)	0.75 ± 0.21 (n=3)	3.8 ± 1.1 (n=3)
Range	0.18 - 0.70	0.17 - 0.71	0.33 - 1.37	0.23 - 1.83	0.72 - 3.32	0.63 - 3.06	0.75 - 3.55	3.1 - 10.1

Table 5. Monthly average air particulate phase concentrations for individual PCB congeners and ∑ 7 PCBs. (Note that for some congener and in some cases the number of samples considered is lower than the samples collected. This is due to analytical problems for those specific congeners in the particular months considered.)

3.3. Seasonal variation

A seasonal trend was observed when firstly plotting total ∑PCB monthly averaged concentration against time (Figure 5). Higher concentrations were observed in summer months whereas lower values were measured in the winter period. Temperature has been described as a key variable affecting PCB ambient concentrations. Gas phase concentration of PCBs seems to exhibit a seasonal trend that is largely the result of cyclical temperature variations, with lower concentrations occurring in the winter months (colder ambient conditions) and higher concentrations in the summer months (warmer conditions) (Manchester-Neesving and Andren, 1989; Hillery et al., 1997; Simcik et al., 1999). Even diurnal cycling has been described for PCBs by some authors (Lee et al., 1998; Hornbuckle and Eisenreich 1996). This phenomenon is most probably due to the higher volatilization rates of PCB from environmental reservoirs when increasing ambient temperature. Volatilization of PCBs from sites were they have been disposed or stored has been identified as an important source (Cousins and Jones, 1998; Simcik et al., 1997).

In our case, the maximum resolution was one week, therefore seasonal cycling was the only phenomenon that could be studied. Weekly integrated concentrations were plotted against weekly average ambient temperature (Figure 6). It seems clear than the PCB air concentration profile follows the atmospheric temperature and that PCBs air concentrations showed a seasonal variation, with higher concentrations in summer and lower concentrations in winter months as described by other authors (see above). The months of April, May and November seem to be a representing a transition situation.

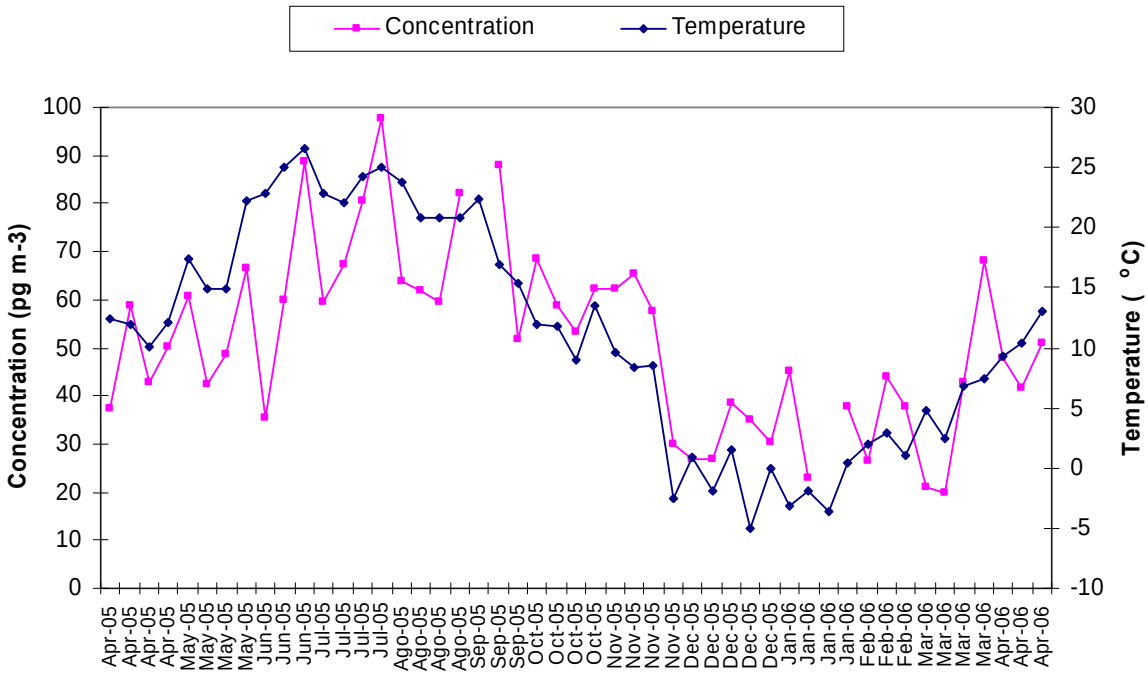


Figure 6. Total ∑7 PCB air concentrations and ambient temperature during the sampling events.

When examining individually the gas phase and the particulate phase ∑PCB concentrations, a positive correlation with temperature was obtained for the gas phase whereas a negative correlation was obtained for the particulate phase concentrations (Figure 7). A positive correlation with temperature was also obtained for the total concentration (gas + particulate phase). This is most probably due to the fact that in general the total concentration was dominated by the gas phase concentrations, especially for the lighter PCBs and only a small contribution from the particulate phase was present. However, for heaviest congener studied (PCB-180), the particulate phase contribution to the total was higher (see section 3.4).

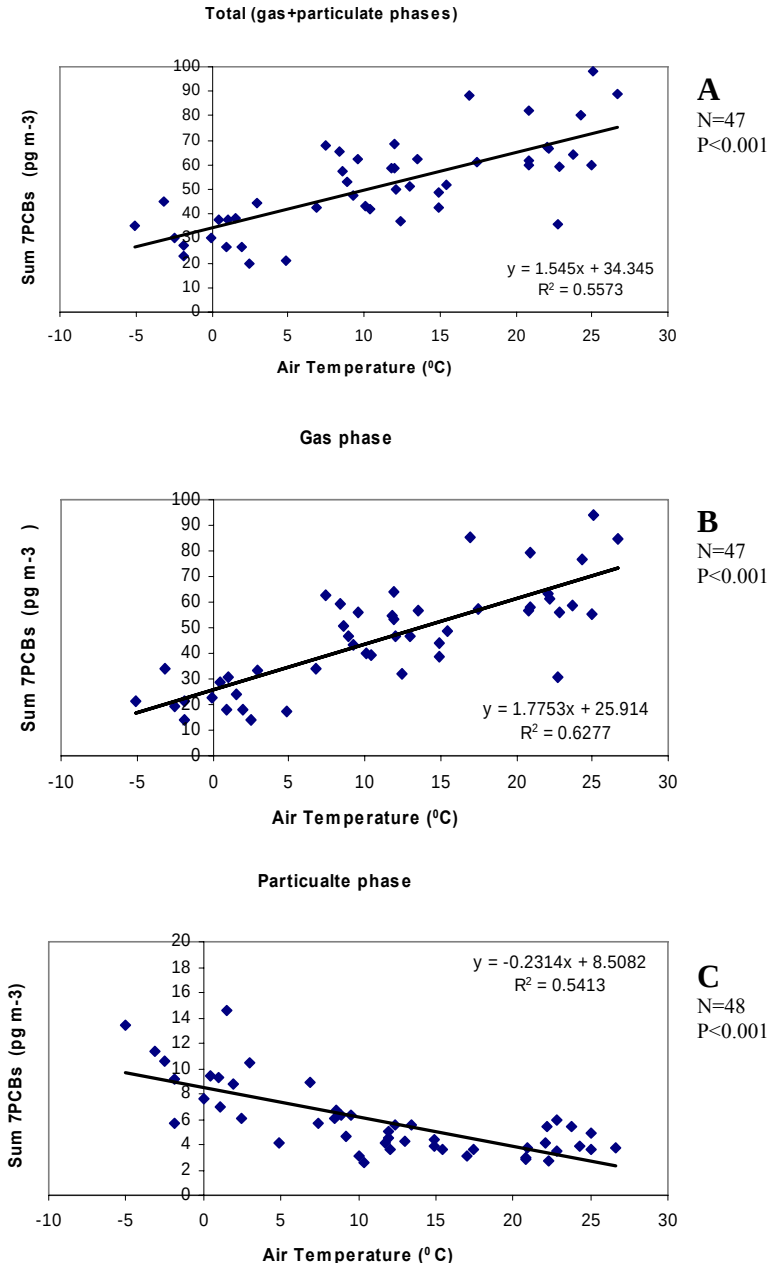


Figure 7. Correlation between ∑7ICES PCB weekly concentrations and ambient temperature for (A) total concentrations (gas + particulate), (B) Gas phase concentrations and (C) particulate phase concentrations

In order to have a better idea on the single PCB congener behaviour, individual congeners were examined. Weekly integrated concentrations were plotted against weekly average ambient temperature for each PCB congener (Annex IV). In addition, linear regression analyses were performed for each of the individuals (Table 6). When examining the results, congeners 52, 101, 118 and 153 seemed to be better correlated with temperature throughout the sampling period, whereas congener 28, 138 and 180 exhibited a weaker correlation.

	TrCB28			TeCB-52			PeCB-101			PeCB-118			HxCB-153			HxCB-138			HpCB-180		
	R^2	N	P	R^2	N	P	R^2	N	P	R^2	N	P	R^2	N	P	R^2	N	P	R^2	N	P
Total	0.196	40	0.004	0.757	42	<0.001	0.724	47	<0.001	0.406	47	<0.001	0.526	47	<0.001	0.363	47	<0.001	0.0696	47	0.073*
Gas Phase	0.214	40	0.003	0.765	42	<0.001	0.754	47	<0.001	0.548	47	<0.001	0.624	47	<0.001	0.486	47	<0.001	0.410	47	<0.001
Particulate Phase	0.167	47	0.004	0.305	47	<0.001	0.558	48	<0.001	0.624	48	<0.001	0.571	48	<0.001	0.493	48	<0.001	0.326	48	<0.001

* not enough power

Table 6. Correlation between individual PCB congener concentration and air temperature during the sampling period.

3.4. Air gas / particulate phase partition and congener patterns

The atmosphere has been identified as a dominant transport pathway for most POPs in the environment. Due to atmospheric transport these pollutant can be moved from sources to distant locations and therefore contaminate remote terrestrial and aquatic environments. A key process determining the fate, transport, atmospheric residence time and removal processes of these chemicals in the atmosphere is their partition between gas and particulate phases (Pankow and Bidleman, 1992; Cotham and Bidleman, 1995).

PCB partition in both air phases is presented in Figure 8. In general, the observed tendency is that the heavier the congener the higher the partition towards the particulate phase. Thus, lighter congeners (CB-28, 52, 101) were more than 90% in the gas phase, whereas the heavier congeners presented smaller contributions to the gas phase (CB-138, 77% and CB-180, 53%). These results are in agreement with previous studies (Hornbuckle et al., 2006; Mandalakis et al., 2002; 2005; Totten et al., 2004; Van Drooge et al., 2002).

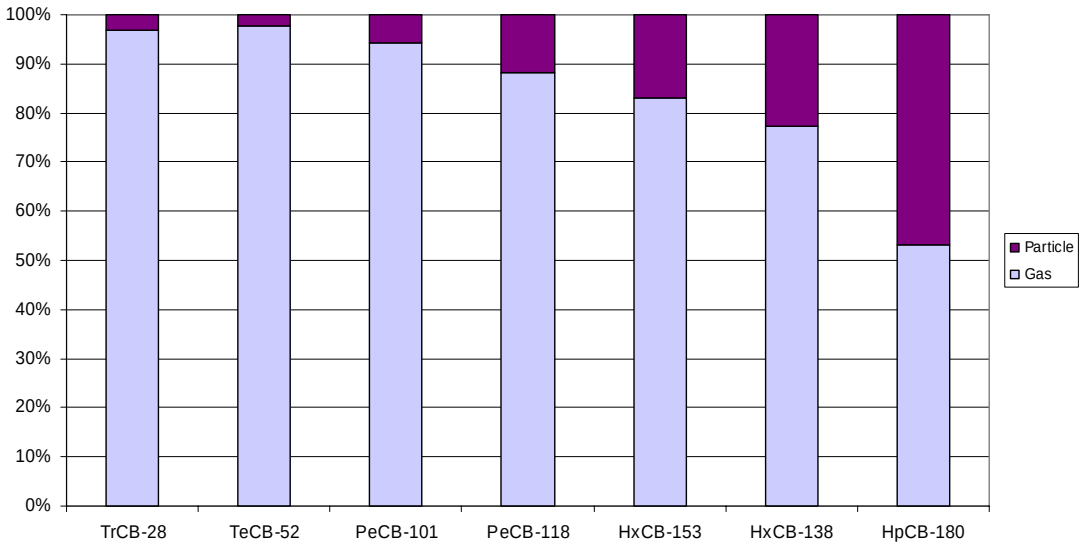


Figure 8. Averaged gas/phase partition of individual PCB congener for the sampling period

PCB congener pattern is depicted in Figure 9. Values are averaged concentrations (gas + particulate phase) throughout the one year sampling period. A predominance of CB-52 was observed. The high standard deviations observed were not only due to the experimental variability but also to the fact of seasonality. As indicated above, concentrations where higher in the high temperature season than in the low temperature period.

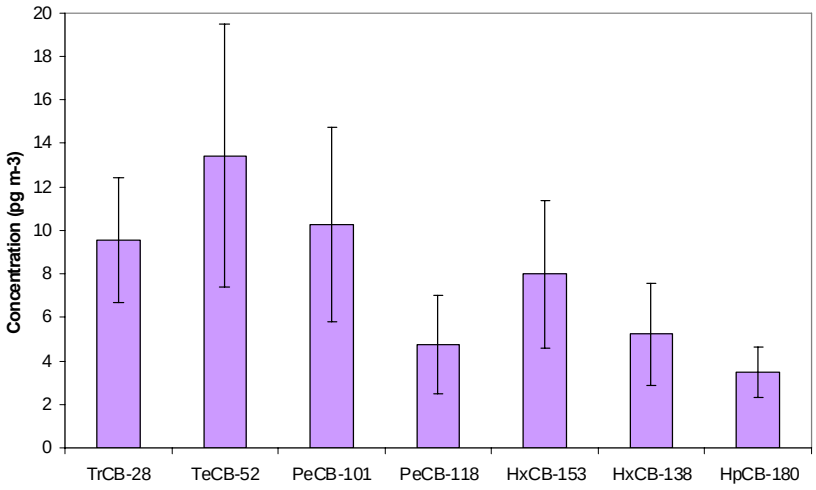


Figure 9. Averaged PCB congener pattern (gas + particulate phase concentrations) obtained for the sampling period. Bars are standard deviation. (TrCB-28, n= 40, TeCB-52, n=42, PeCB-101, n=47, PeCB-118, n=47, HxCB-153, n=46, HxCB-138, n=46, HpCB-180, n=47).

In order to investigate whether the pattern was consistent in the warmer and the colder periods, the concentrations corresponding to the 13 highest or lowest (depending of the period considered) weekly average temperatures were only considered. Results are presented in Figure 10.

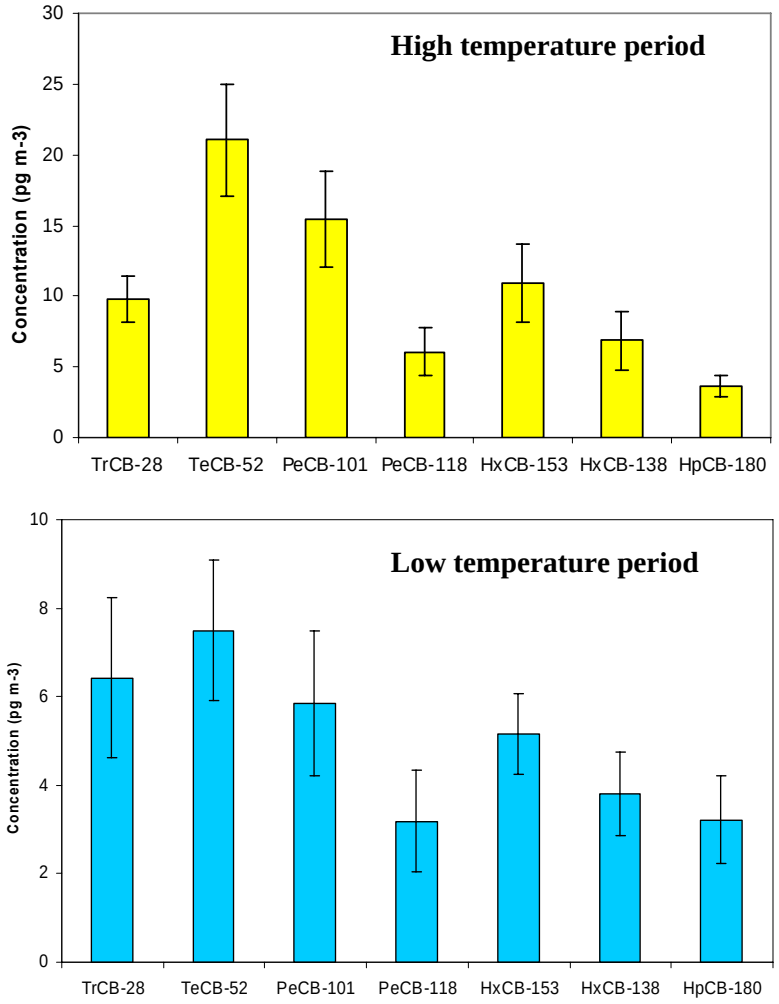


Figure 10. Averaged PCB congener pattern (gas + particulate phase concentrations) obtained for the highest and lowest temperature periods. Bars are standard deviation. (TrCB-28, n= 10, TeCB-52, n=11, PeCB-101, n=12, PeCB-118, n=12, HxCB-153, n=12, HxCB-138, n=12, HpCB-180, n=12.

Standard deviations were generally lower, as expected. The obtained patterns were almost identical for the higher chlorinated congeners in both periods, but some sort of variation was observed on the left side of the graph. The predominance of the CB-52 was not as clear for the colder period since the CB-28 gained importance.

PCB congener patterns in the air gas and particulate phases were also studied for both periods. Results are presented in Figure 11. Pattern observed in the gas phase were almost identical to the ones found for the total concentration (Figure 10) supporting the observation of the higher partition to the gas phase, especially for the lighter congeners. However, a predominance of the higher chlorinated

congeners was observed in the particular gas phase, and almost identical signal were obtained for both periods.

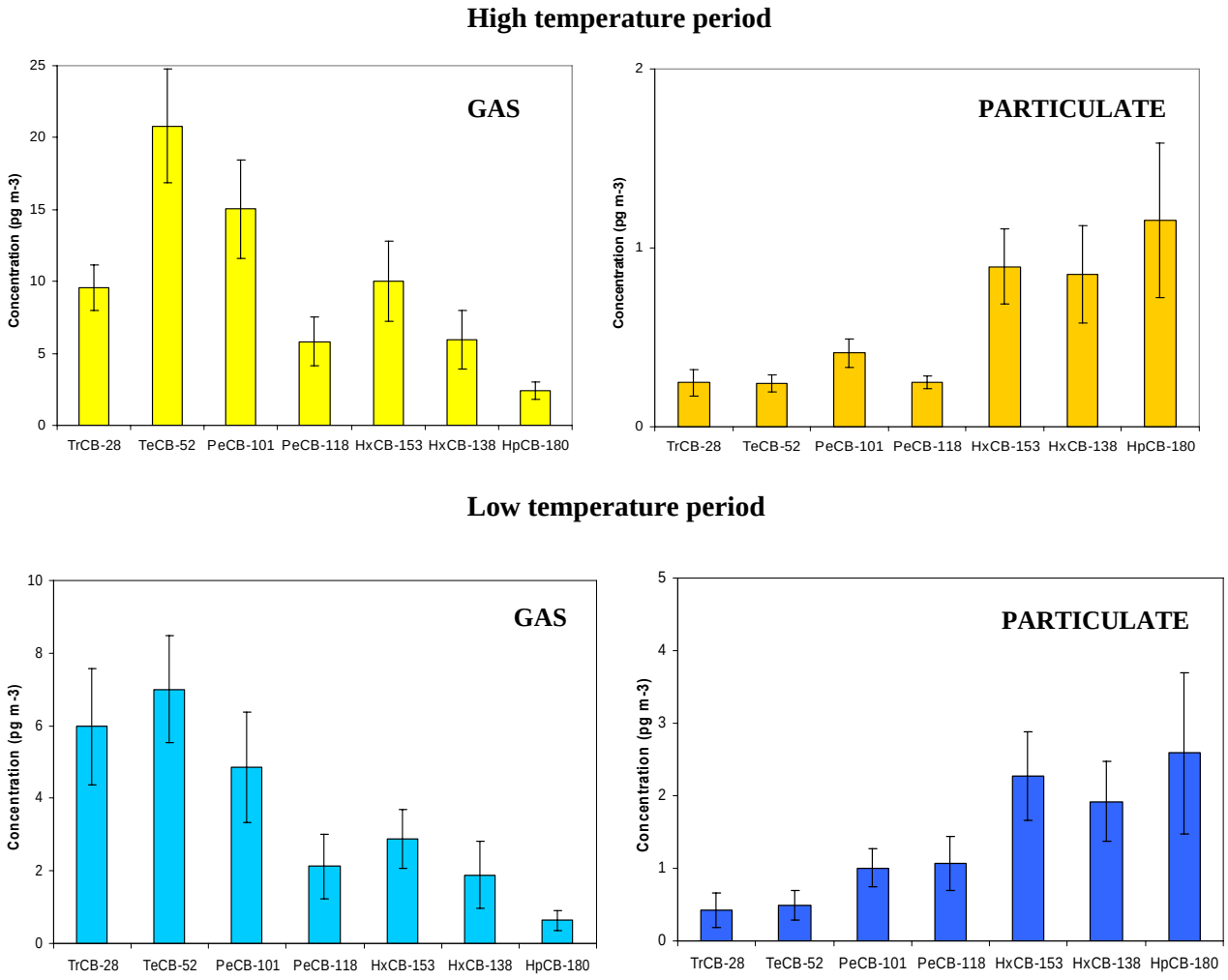


Figure 11. Averaged PCB congener pattern in the air gas and particulate phases obtained for the sampling period. Bars are standard deviation (TrCB-28, n= 10-13, TeCB-52, n=10-13, PeCB-101, n=12-13, PeCB-118, n=12-13, HxCB-153, n=12-13, HxCB-138, n=12-13, HpCB-180, n=12-13. note that "n" varied depending on individual congeners and phases).

However, is has to be taken into account that losses of the most volatile congeners, probably due to excessive sample extract evaporation, were suspected for some samples, and CB-28 was the most affected. For this congener, the analytical recoveries were very low in several samples and this could be also affecting the pattern found.

Another interesting observation was the effect of the seasonality on the gas-particulate phase partition for individual congeners. The percentages of concentrations found in the gas phase for each congener in both periods are presented in Figure 12.

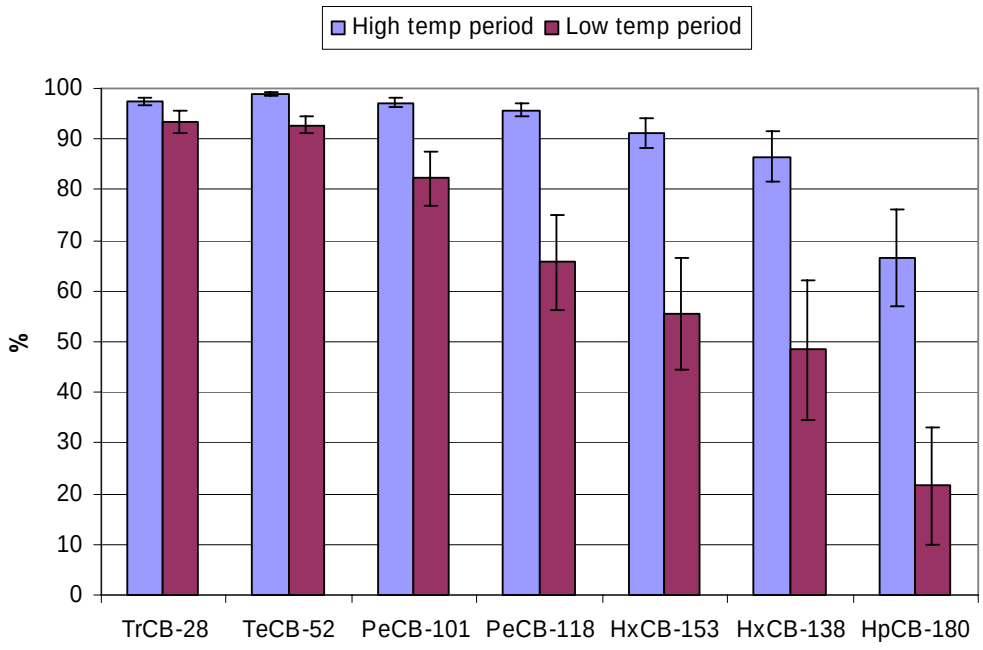


Figure 12. Percentages of individual PCB congeners in the air gas phase for the high and low temperature periods. Bars are relative standard deviations (TrCB-28, n= 10, TeCB-52, n=10, PeCB-101, n=12, PeCB-118, n=12, HxCB-153, n=12, HxCB-138, n=12, HpCB-180, n=12).

It can be seen that in general the percentage in the gas phase is lower for the low temperature season than for the warmer period. In addition, this effect seems to be of different magnitude depending on the PCB congener, being remarkably more noticed for the higher chlorinated congeners (right side of graph). This observation is in agreement with the findings of other authors (Halsall et al., 1995).

4. Final remarks

Ambient air concentrations for the 7 indicators PCB (28, 52, 101, 118, 153, 138, and 180) have been reported for the first time in the sub-alpine location of Ispra (JRC Ispra site). Concentrations are within the range of those reported for rural, semi-rural or remote areas around the world. A seasonal variation of air concentrations was observed, with higher levels in summer months (higher average temperatures) and lower values in winter (lower average temperatures). In addition, indications of seasonal variation affecting congener patterns and the gas - particulate phase partition for the studied PCB were found. PCBs were in general predominant in the air gas phase, dominating therefore the contribution to the total airborne concentration, although the percentage in the gas phase decreases when increasing level of chlorination and during the low temperature season (winter months).

A one year data set of PCB ambient air concentrations has been generated constituting a useful dataset that now remains available for further use, such as inclusion in the EMEP network or for POPs fate modelling validation and future risk assessment of POPs in sub-alpine locations. More results for other target POPs are under processing and are expected to be reported soon.

5. Acknowledgements

We would like to thank M. Ghiani for her help with the sampling and E. Christoph for his help with sampling and his assistance with the clean-up system. We would like to acknowledge A. Dell'Acqua for his support on the setting up of the sampling POPs site at the EMEP Station and to F. Lagler for his support with the air sampling equipment and necessary material.

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7. Annexes

7.1. Annex I: Air sampling overview: April 2005-March 2007.

MONTH	SAMPLE code	Start date	Finish date	Flow- l/min -	Vol (m^3)	Atm pressure (kPa)*	At. Av T (^oC)*
Apr-05	EMEP-2	30-Mar-05	6-Apr-05	86.980	869.216	99.870	12.4
Apr-05	EMEP-3	6-Apr-05	13-Apr-05	86.989	864.515	98.667	12.0
Apr-05	EMEP-4	14-Apr-05	21-Apr-05	86.984	869.270	97.254	10.1
Apr-05	EMEP-5	21-Apr-05	28-Apr-05	86.988	834.926	98.515	12.1
May-05	EMEP-6	29-Apr-05	6-May-05	86.986	856.338	98.692	17.5
May-05	EMEP-7	6-May-05	13-May-05	86.987	836.364	98.348	14.9
May-05	EMEP-8	13-May-05	20-May-05	86.987	848.821	98.579	15.0
May-05	EMEP-9	23-May-05	30-May-05	86.989	869.203	99.543	22.2
Jun-05	EMEP-10	30-May-05	6-Jun-05	86.988	868.880	99.179	22.8
Jun-05	EMEP-11	15-Jun-05	22-Jun-05	86.989	869.146	99.477	25.0
Jun-05	EMEP-12	22-Jun-05	30-Jun-05	86.989	869.249	99.211	26.6
Jul-05	EMEP-13	30-Jun-05	7-Jul-05	85.479	854.153	98.617	22.9
Jul-05	EMEP-14	8-Jul-05	15-Jul-05	86.988	869.179	99.053	22.1
Jul-05	EMEP-15	15-Jul-05	22-Jul-05	86.988	869.213	98.841	24.3
Jul-05	EMEP-16	22-Jul-05	29-Jul-05	86.990	869.125	98.870	25.0
Ago-05	EMEP-17	29-Jul-05	5-Aug-05	86.990	869.239	98.656	23.7
Ago-05	EMEP-18	5-Aug-05	11-Aug-05	102.800	876.517	98.798	20.9
Ago-05	EMEP-19	11-Aug-05	18-Aug-05	86.988	869.257	98.615	20.8
Ago-05	EMEP-21	24-Aug-05	31-Aug-05	86.988	869.241	99.070	20.9
Sep-05	EMEP-22	31-Aug-05	7-Sep-05	86.989	869.281	99.323	22.3
Sep-05	EMEP-23	7-Sep-05	14-Sep-05	86.990	869.288	98.936	17.0
Sep-05	EMEP-25	21-Sep-05	28-Sep-05	86.991	869.294	99.221	15.4
Oct-05	EMEP-26	28-Sep-05	5-Oct-05	86.990	869.289	99.054	12.0
Oct-05	EMEP-27	5-Oct-05	12-Oct-05	86.990	869.286	99.568	11.8
Oct-05	EMEP-28	12-Oct-05	19-Oct-05	86.990	869.262	99.590	8.9
Oct-05	EMEP-29	20-Oct-05	28-Oct-05	88.470	861.885	99.139	13.5
Nov-05	EMEP-30	28-Oct-05	4-Nov-05	86.990	869.291	99.492	9.6
Nov-05	EMEP-31	4-Nov-05	11-Nov-05	86.989	869.059	99.789	8.4
Nov-05	EMEP-32	11-Nov-05	18-Nov-05	86.988	869.262	98.526	8.6
Nov-05	EMEP-33	18-Nov-05	25-Nov-05	86.990	869.312	98.911	-2.5
Dec-05	EMEP-34	25-Nov-05	2-Dec-05	86.989	869.302	97.249	1.0
Dec-05	EMEP-35	2-Dec-05	9-Dec-05	88.654	872.815	97.934	-1.9
Dec-05	EMEP-36	9-Nov-05	16-Dec-05	86.988	869.276	99.154	1.5
Dec-05	EMEP-37	16-Dec-05	23-Dec-05	86.990	852.411	98.760	-5.1
Dec-05	EMEP-38	23-Dec-05	30-Dec-05	86.989	869.273	97.969	0.0
Jan-06	EMEP-39	3-Jan-06	10-Jan-06	86.991	869.192	99.183	-3.2
Jan-06	EMEP-40	10-Jan-06	17-Jan-06	86.986	869.153	99.598	-1.9
Jan-06	EMEP-41	17-Jan-06	24-Jan-06	86.933	703.083	98.745	-3.7
Jan-06	EMEP-42	24-Jan-06	31-Jan-06	86.988	869.263	99.085	0.5
Feb-06	EMEP-44	7-Feb-06	14-Feb-06	86.989	869.330	98.392	2.0
Feb-06	EMEP-45	14-Feb-06	21-Feb-06	86.989	869.197	97.699	3.0
Feb-06	EMEP-46	21-Feb-06	28-Feb-06	86.99	864.4	98.014	1.1
Mar-06	EMEP-47	28-Feb-06	7-Mar-06	86.989	869.252	97.344	4.8
Mar-06	EMEP-48	7-Mar-06	14-Mar-06	86.991	869.328	98.055	2.5
Mar-06	EMEP-49	14-Mar-06	21-Mar-06	86.988	869.267	98.39	6.8
Mar-06	EMEP-50	21-Mar-06	28-Mar-06	86.989	788.495	98.286	7.5
Apr-06	EMEP-51	28-Mar-06	4-Apr-06	86.989	869.181	98.636	9.3
Apr-06	EMEP-52	4-Apr-06	11-Apr-06	86.989	869.299	98.009	10.4
Apr-06	EMEP-53	11-Apr-06	18-Apr-06	86.989	869.223	98.366	13.0
Apr-06	EMEP-54	18-Apr-06	25-Apr-06	86.990	869.250	98.793	13.36
May-06	EMEP-55	26-Apr-06	3-May-06	86.989	868.825	98.375	15.87
May-06	EMEP-56	3-May-06	10-May-06	86.989	869.262	98.996	16.63
May-06	EMEP-57	10-May-06	17-May-06	86.988	869.276	99.076	18.92
May-06	EMEP-58	17-May-06	24-May-06	86.988	869.060	98.565	19.96
May-06	EMEP-59	24-May-06	31-May-06	86.988	868.994	98.750	19.47
Jun-06	EMEP-60	31-May-06	7-Jun-06	86.988	869.315	99.042	19.71
Jun-06	EMEP-61	7-Jun-06	14-Jun-06	86.988	869.206	99.601	21.58
Jun-06	EMEP-62	14-Jun-06	21-Jun-06	86.988	869.281	99.244	24.92
Jun-06	EMEP-63	21-Jun-06	28-Jun-06	86.988	868.981	98.966	26.75
Jul-06	EMEP-64	28-Jun-06	5-Jul-06	86.988	869.299	99.173	26.47
Jul-06	EMEP-65	06-Jul-06	13-Jul-06	86.988	869.270	99.347	24.98
Jul-06	EMEP-66	13-Jul-06	20-Jul-06	86.990	869.346	99.690	24.13
Jul-06	EMEP-67	20-Jul-06	27-Jul-06	86.988	869.265	99.255	24.7
Ago-06	EMEP-68	27-Jul-06	3-Aug-06	86.989	868.963	98.594	22.47
Ago-06	EMEP-69	03-Aug-06	10-Aug-06	86.988	869.242	98.462	19.45
Ago-06	EMEP-70	10-Aug-06	17-Aug-06	86.989	869.283	98.477	16.04
Ago-06	EMEP-71	17-Aug-06	24-Aug-06	86.988	869.265	99.043	19.25
Ago-06	EMEP-72	24-Aug-06	31-Aug-06	86.988	869.166	98.554	17.19
Sep-06	EMEP-73	31-Aug-06	7-Sep-06	86.988	869.168	99.533	22.29
Sep-06	EMEP-74	07-Sep-06	11-Sep-06	142.820	830.794	99.636	18.94
Sep-06	EMEP-75	13-Sep-06	20-Sep-06	86.987	820.776	98.614	15.87
Sep-06	EMEP-76	20-Sep-06	27-Sep-06	86.989	869.284	98.993	16.04
Oct-06	EMEP-77	27-Sep-06	4-Oct-06	86.988	868.869	98.798	16.15
Oct-06	EMEP-78	4-Oct-06	11-Oct-06	86.987	863.411	99.354	11.85
Oct-06	EMEP-79	12-Oct-06	19-Oct-06	86.988	869.031	99.566	11.78
Oct-06	EMEP-80	19-Oct-06	26-Oct-06	86.988	869.132	98.629	12.35
Nov-06	EMEP-81	26-Oct-06	6-Nov-06	86.988	869.250	99.189	11.52
Nov-06	EMEP-82	6-Nov-06	13-Nov-06	86.988	869.236	99.679	6.64
Nov-06	EMEP-83	13-Nov-06	20-Nov-06	86.988	869.312	99.384	9.12
Nov-06	EMEP-84	20-Nov-06	27-Nov-06	86.989	869.260	98.735	7.56
Dec-06	EMEP-85	27-Nov-06	4-Dec-06	86.988	869.309	100.020	6.11
Dec-06	EMEP-86	5-Dec-06	12-Dec-06	86.989	869.270	99.044	5.82
Dec-06	EMEP-87	12-Dec-06	19-Dec-06	86.988	869.304	100.050	3.11
Dec-06	EMEP-88	19-Dec-06	26-Dec-06	86.988	869.184	100.410	-0.12
Jan-07	EMEP-89	09-Jan-07	16-Jan-07	86.986	869.086	99.835	7.74
Jan-07	EMEP-90	16-Jan-07	23-Jan-07	86.988	869.244	99.208	5.87
Jan-07	EMEP-91	23-Jan-07	30-Jan-07	86.989	869.263	98.458	5.28
Feb-07	EMEP-92	30-Jan-07	6-Feb-07	86.989	869.106	99.330	5.90
Feb-07	EMEP-93	6-Feb-07	13-Feb-07	86.989	869.320	97.825	7.67
Feb-07	EMEP-94	13-Feb-07	20-Feb-07	86.987	759.487	98.897	7.82
Feb-07	EMEP-95	20-Feb-07	27-Feb-07	86.988	869.283	98.413	6
Mar-07	EMEP-96	27-Feb-07	6-Mar-07	86.988	868.924	98.532	8.12
Mar-07	EMEP-97	07-Mar-07	14-Mar-07	86.989	869.257	99.460	8.49
Mar-07	EMEP-98	14-Mar-07	21-Mar-07	86.988	869.280	98.201	9.9
Mar-07	EMEP-99	21-Mar-07	28-Mar-07	86.989	869.145	98.315	7.14

: SAMPLING PERFORMED IN DUPLICATE (QA/QC)

* Averaged atmopsheric pressure and temperature during the sampling event. Values were recorded by the meteo sensor from the air sampler

7.2. Annex II: Precipitation sampling overview: January 2005-December 2006

SAMPLED SEASON	SAMPLE	Start day	Finish date
Winter 05	P-1	25-Jan-05	7-Mar-05
Spring 05	P-2	22-Mar-05	30-Mar-05
Spring 05	P-3	30-Mar-05	19-Apr-05
Spring /Summer 05	P-4	19-Apr-05	19-Ago-05
Summer/ Autum 05	P-5	19-Ago-05	12-Oct-05
Autum 05	P-6	12-Oct-05	6-Dec-05
Winter 05- 06	P-7	6-Dec-05	14-Feb-06
Winter 06	P-8	14-Feb-06	9-Mar-06
Spring 06	P-9	9-Mar-06	10-Apr-06
Spring 06	P-10	10-Apr-06	10-May-06
Spring /Summer 06	P-11	10-May-06	10-Jun-06
Summer 06	P-12	10-Jun-06	17-Ago-06
Summer /Autum 06	P-13	17-Ago-06	7-Nov-06
Winter 06	P-14	7-Nov-06	22-Dec-06

7.3. Annex III: List of target POPs

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs)

2,3,7,8-TCDD
1,2,3,7,8-PeCDD
1,2,3,4,7,8-HxCDD
1,2,3,6,7,8-HxCDD
1,2,3,7,8,9-HxCDD
1,2,3,4,6,7,8-HpCDD
OCDD
2,3,7,8-TCDF
1,2,3,7,8-PeCDF
2,3,4,7,8-PeCDF
1,2,3,4,7,8-HxCDF
1,2,3,6,7,8-HxCDF
2,3,4,6,7,8-HxCDF
1,2,3,7,8,9-HxCDF
1,2,3,4,6,7,8-HpCDF
1,2,3,4,7,8,9-HpCDF
OCDF

Polychlorinated biphenyls (PCBs)

Indicators
TrCB28
TeCB-52
PeCB-101
PeCB-118
HxCB-153
HxCB-138
HpCB-180
Dioxin-like PCBs
TeCB-81
TeCB-77
PeCB-126
HxCB-169
PeCB-105
PeCB-114
PeCB-118
PeCB-123
HxCB-156
HxCB-157
HxCB-167
HpCB-189

Polybrominated diphenyl ethers (PBDEs)

BDE-17
BDE-28
BDE-47
BDE-49
BDE-66
BDE-85
BDE-99
BDE-100
BDE-153
BDE-154
BDE-183
BDE-196
BDE-197
BDE-203

BDE-206
BDE-207
BDE-208
BDE-209

Organochlorine pesticides (OCPs)

HCB
Alpha-HCH
Beta-HCH
Gamma-HCH (Lindane)
Heptachlor
Heptachlor-exo-epoxide (cis)
Heptachlor-endo- epoxide (trans)
Aldrin
Dieldrin
Endrin
Oxychlordane
Cis-chlordane
Trans-chlordane (gamma)
Trans-nonachlor
Cis-nonachlor
2,4' DDE
4,4' DDE
2,4' DDD
4,4' DDD
2,4' DDT
4,4' DDT
Endosulfan-alpha
Endosulfan-beta
Endosulfan-sulfate
Mirex

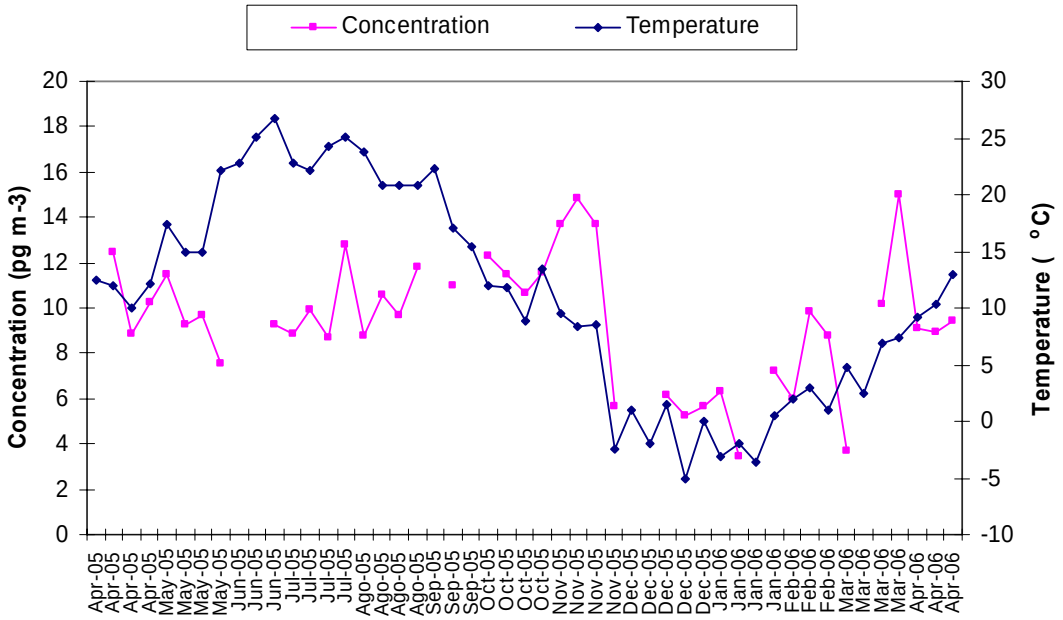
Polycyclic aromatic hydrocarbons (PAHs)

Napthalene
Acenapthylene
Acenapthene
Fluorene
Phenantrhene
Anthracene
Fluoranthene
Pyrene
Benzo (a) anthracene
Chrysene
Benzo(b+k)fluoranthene
Benzo (a) pyrene
Indene (1,2,3-cd) pyrene
Dibenzo (a, h) anthracene
Benzo (g, h, i) perylene

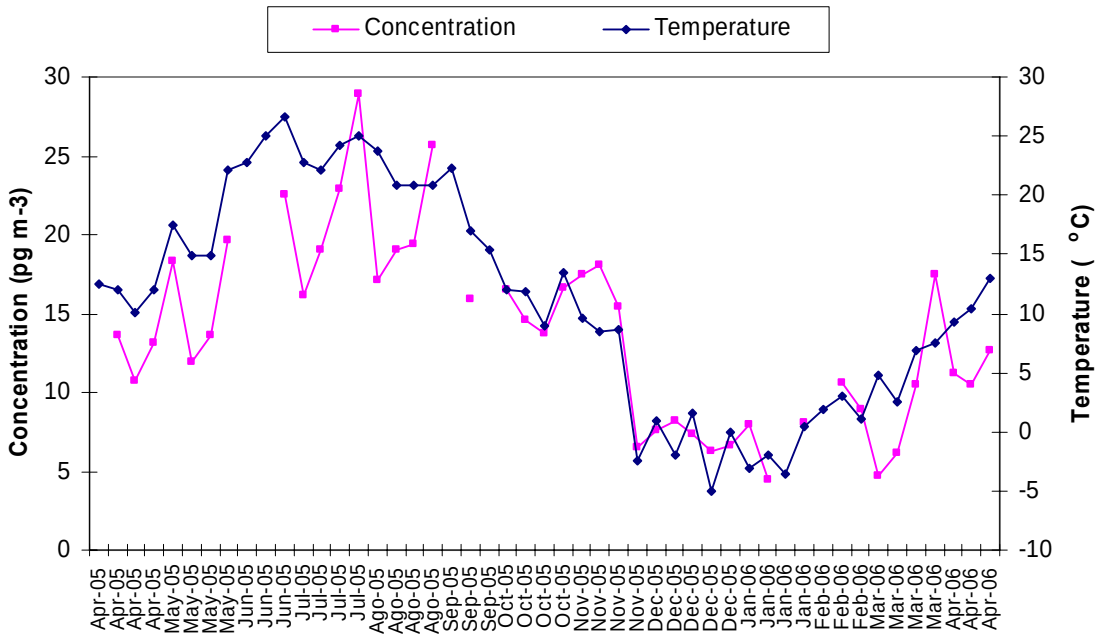
7.4. Annex IV. Individual PCB congener plots (concentration & temperature)

Note: Concentrations displayed are total concentrations (gas + particulate phase)

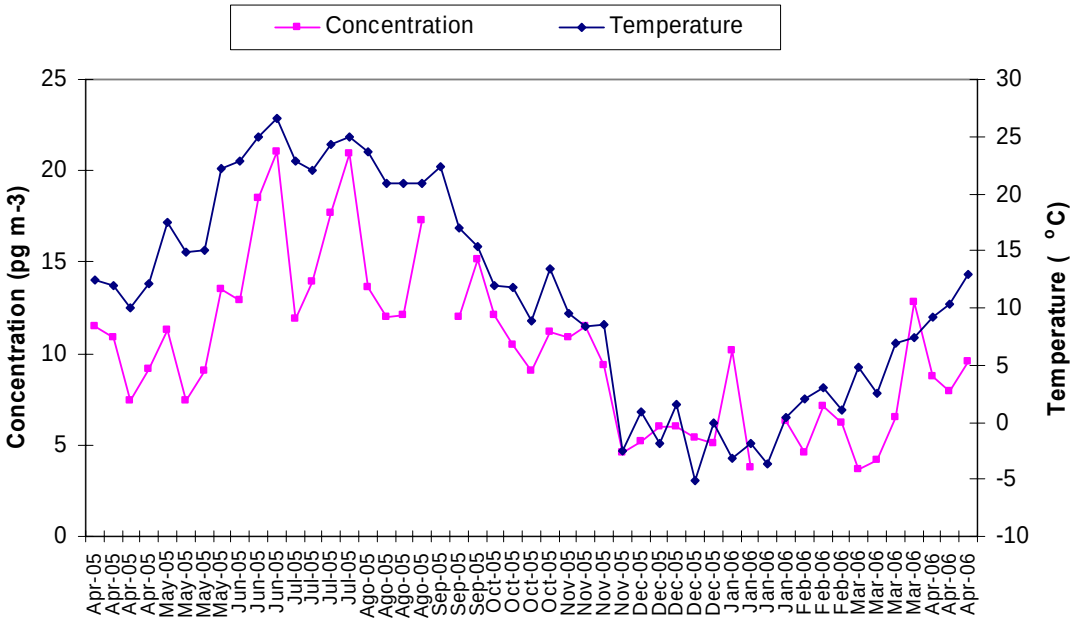
CB-28



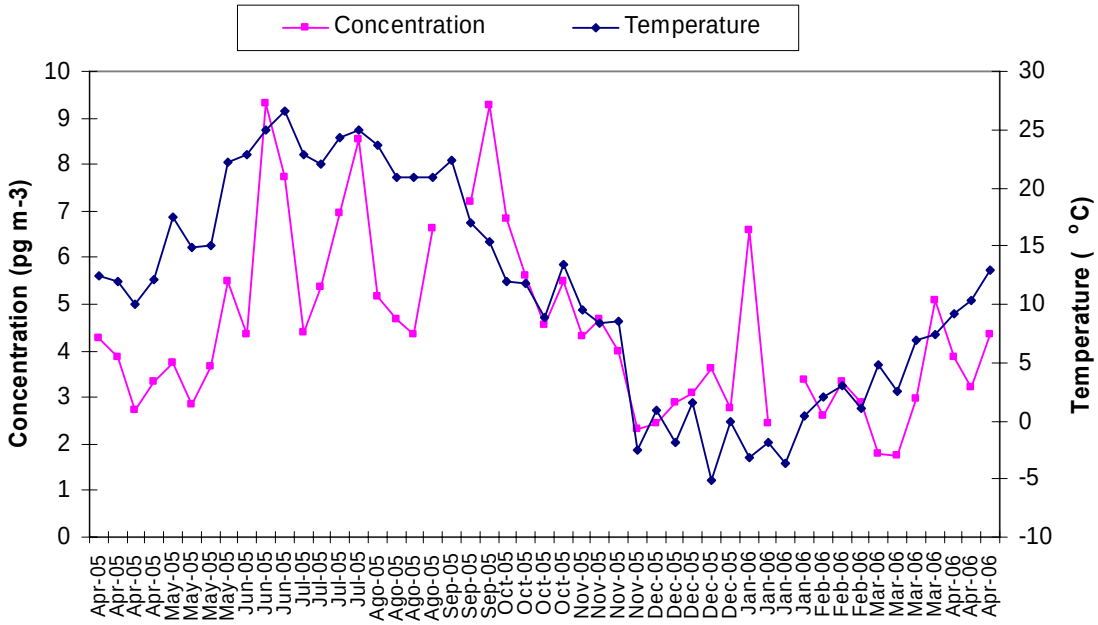
CB-52



CB-101


Concentration
Temperature
Concentration (pg m-3)
Temperature (°C)
0
5
10
15
20
25
-10
-5
0
5
10
15
20
25
30
Apr-05
Apr-05
Apr-05
Apr-05
May-05
May-05
May-05
May-05
Jun-05
Jun-05
Jun-05
Jul-05
Jul-05
Jul-05
Jul-05
Ago-05
Ago-05
Ago-05
Ago-05
Sep-05
Sep-05
Sep-05
Oct-05
Oct-05
Oct-05
Oct-05
Nov-05
Nov-05
Nov-05
Nov-05
Dec-05
Dec-05
Dec-05
Dec-05
Dec-05
Jan-06
Jan-06
Jan-06
Feb-06
Feb-06
Feb-06
Mar-06
Mar-06
Mar-06
Mar-06
Apr-06
Apr-06
Apr-06

CB-118


Concentration
Temperature
Concentration (pg m-3)
Temperature (°C)
0
1
2
3
4
5
6
7
8
9
10
-10
-5
0
5
10
15
20
25
30
Apr-05
Apr-05
Apr-05
Apr-05
May-05
May-05
May-05
May-05
Jun-05
Jun-05
Jun-05
Jul-05
Jul-05
Jul-05
Jul-05
Ago-05
Ago-05
Ago-05
Ago-05
Sep-05
Sep-05
Sep-05
Oct-05
Oct-05
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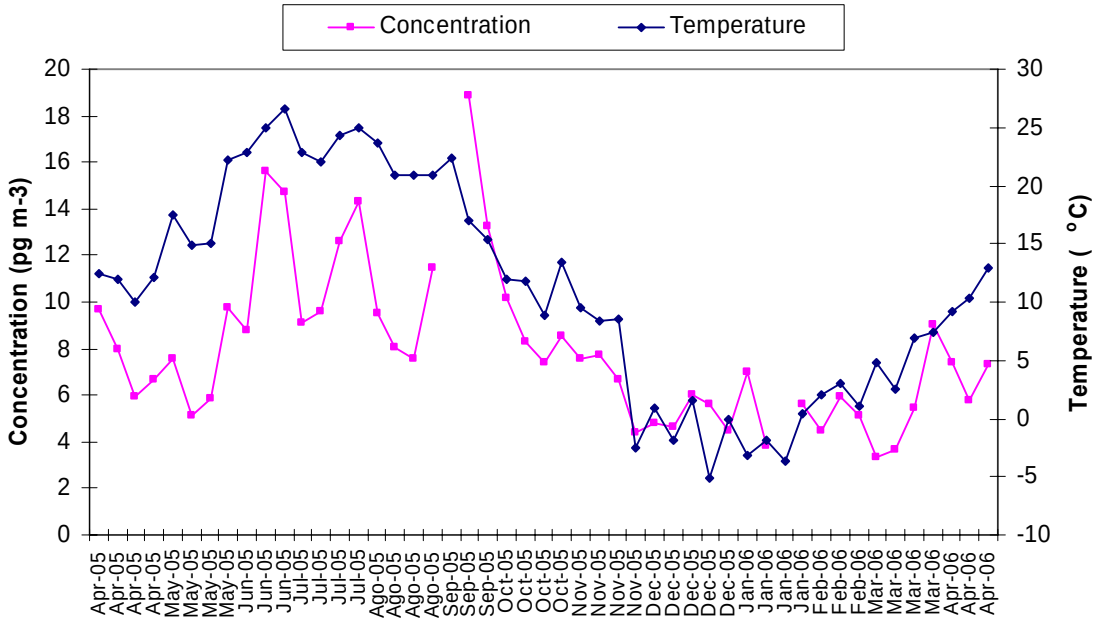
Apr-06
Apr-06
Apr-06

CB-153


Concentration
Temperature
Concentration (pg m-3)
Temperature (°C)
0
2
4
6
8
10
12
14
16
18
20
-10
-5
0
5
10
15
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Apr-05
Apr-05
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Apr-05
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Jul-05
Jul-05
Jul-05
Jul-05
Ago-05
Ago-05
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Ago-05
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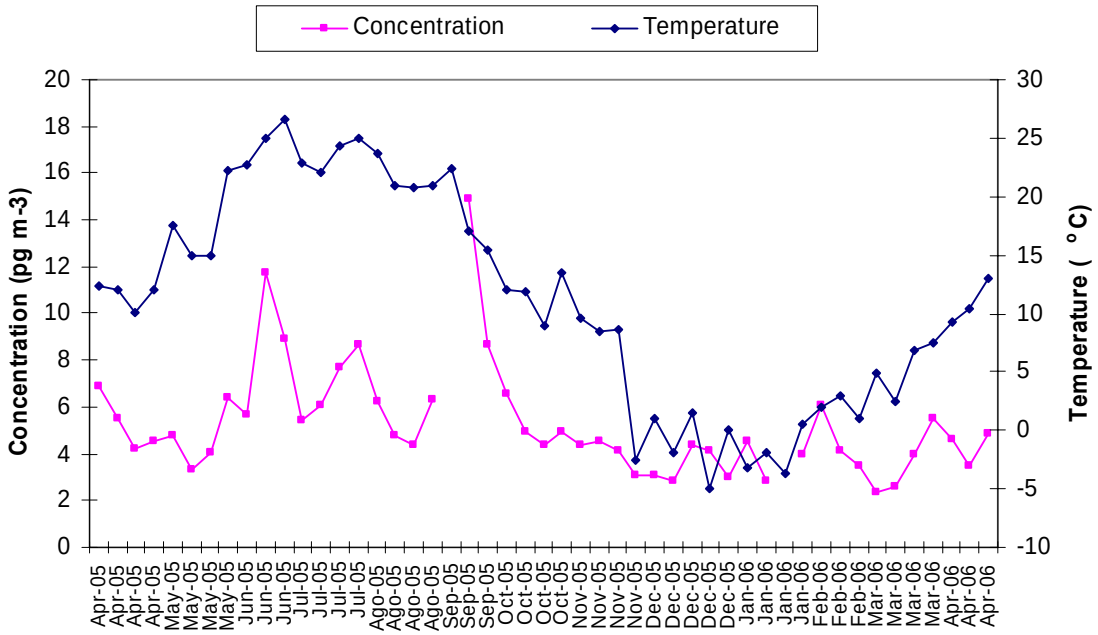
Apr-06
Apr-06

CB-138


Concentration
Temperature
Concentration (pg m-3)
Temperature (°C)
0
2
4
6
8
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12
14
16
18
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-10
-5
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Jul-05
Ago-05
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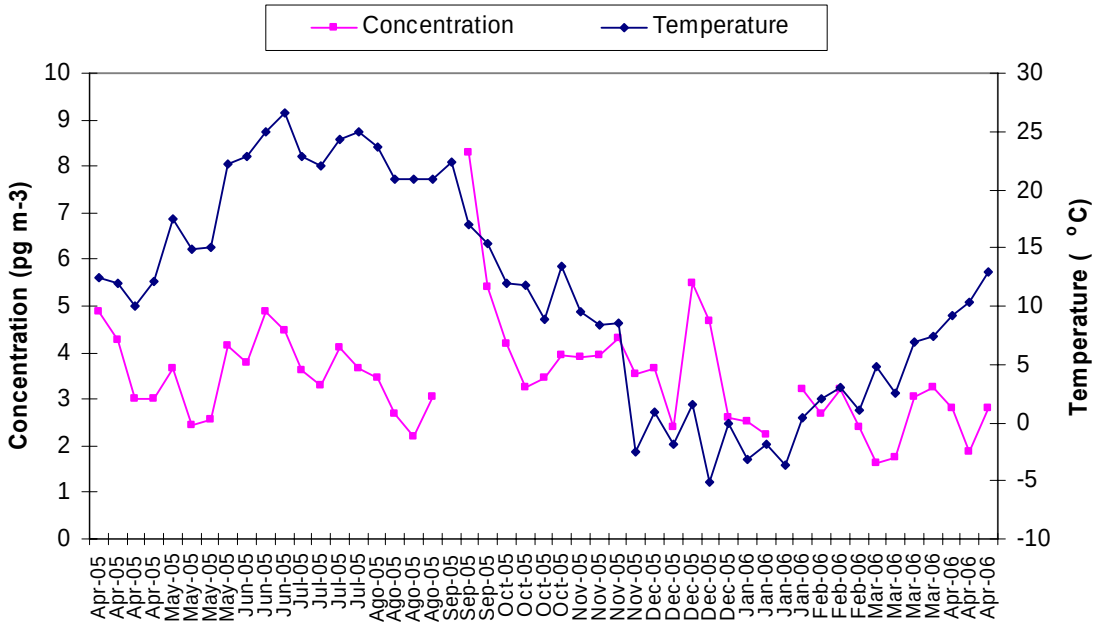
CB-180


Concentration
Temperature
Concentration (pg m-3)
Temperature (°C)
0
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Apr-06

European Commission

EUR 23258 EN – Joint Research Centre – Institute for Environment and Sustainability
Title: Polychlorinated biphenyls (PCBs) at the JRC Ispra Site: Air Concentrations, Congener Patterns and Seasonal variation. - Results from the 1st year of atmospheric monitoring of persistent organic pollutants (POPs) at the Ispra EMEP station

Author(s): J. Castro-Jiménez, S.J. Eisenreich, G. Mariani, H. Skejo, and G. Umlauf

Luxembourg: Office for Official Publications of the European Communities
2008 – 38 pp.
EUR – Scientific and Technical Research series – ISSN 1018-5593

Abstract
With the aim of gathering information on atmospheric POPs concentrations in one of the "monitoring holes" in Europe, a monitoring and research atmospheric site was set up at the JRC Ispra Site. Regular monitoring activities started on April 2005 and finished on March 2007 in this first stage. Air and precipitation samples were collected regularly throughout this period. First results obtained on PCBs ambient air concentrations (7 indicator PCBs: 28, 52, 101, 118, 153, 138 and 180) from the first year of monitoring (April 2005-2006) are presented in this report. In addition, congener patterns, air gas/particulate phase partition and seasonal variation are discussed. The range of ∑7 PCB total (gas + particulate phases) monthly averaged concentration during the sampling period varied from 76 ± 17 to 31 ± 5 pg m^{-3}. Concentrations are within the range of those reported for rural, semi-rural or remote areas around the world. A seasonal variation of air concentrations was observed, with higher levels in summer months (higher average temperatures) and lower values in winter (lower average temperatures). In addition, indications of seasonal variation affecting congener patterns and the gas - particulate phase partition for the studied PCB were found. PCBs were in general predominant in the air gas phase, dominating therefore the contribution to the total airborne concentration, although the percentage in the gas phase decreases when increasing level of chlorination and during the low temperature season (winter months).

A one year data set of PCB ambient air concentrations has been generated, constituting a useful dataset that now remains available for further use, such as inclusion in the EMEP network or for POPs fate modelling validation and future risk assessment of POPs in sub-alpine locations. More results for other target POPs are under processing and are expected to be reported soon.

The mission of the JRC is to provide customer-driven scientific and technical support for the conception, development, implementation and monitoring of EU policies. As a service of the European Commission, the JRC functions as a reference centre of science and technology for the Union. Close to the policy-making process, it serves the common interest of the Member States, while being independent of special interests, whether private or national.